Chapter 3
Novel Strategies for Structuring Liquid Oils, Their Applications, and Health Implications

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Abbreviations

12-HAS	12-hydoxystearic acid
A_w	Water activity
BW	Beeswax
CLW	Candelilla wax
CRW	Carnauba wax
CVD	Cardiovascular disease
DAGs	Diacylglycerols
EC	Ethyl-cellulose
FDA	Food and Drug Administration
GL	Glycerolysis
GP	Glycerolysis product
HDL	High-density lipoprotein
HIPE	High-internal phase emulsion
HLB	Hydrophilic-lipophilic balance
HOAO	High oleic algal
HOCO	High oleic canola
HOSO	High oleic sunflower oil
HPMC	Hydroxypropyl methylcellulose
LDL	Low-density lipoprotein
LMOG	Low molecular weight gelators
MAGs	Monoacylglycerols

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MC	Methylcellulose
PHOs	Partially hydrogenated oils
RBW	Rice bran wax
SFC	Solid fat content
SFW	Sunflower wax
SLC	Starch-lipid composites
SOSA	Stearyl alcohol/stearic acid
TAGs	Triacylglycerol
T_g	Glass transition temperature
WHO	World Health Organization
XG	Xanthan gum

3.1 Introduction

Edible fats are composed of triacylglycerol molecules (TAGs). These TAGs consist of a glycerol backbone with three fatty acid chains attached by ester linkages. The chemical nature of the fatty acids gives a solid-like or liquid-like character to a particular fat at room temperature, and the interaction between polycrystalline TAG colloids is what traditionally structures fat [1–5]. Typically, a bulk fat containing a large proportion of long-chain fatty acids, with a high degree of saturation or high content of *trans* unsaturated fatty acids, results in fat with a higher melting point that is solid at room temperature. In contrast, fats containing high amounts of short-chain fatty acids, or fatty acids with a high degree of *cis* unsaturation, have lower melting points and are liquid at room temperature. This is the main difference between fats and oils, and what dictates the suitability of fat in a particular food application. Semi-solid fats provide a variety of characteristics to different baked goods that oils cannot. For example, solid fats can trap small air bubbles that stay in cake batters longer, producing more reliably fluffy cakes. Solid fats can also keep dough layers apart in pastries resulting in increased volume or lift, whereas, using oil for these applications would result in lower-volume cakes and dough layers that adhere together and leak oil [6]. As the reader might have noticed, fat functionality in many industrial applications requires high contents of saturated or *trans* fat. This functional requirement, however, goes against most health recommendations.

The health effects of fat consumption and the associated risk of cardiovascular disease (CVD) have been a controversial area of research over the past several decades [7–10]. The health controversy surrounding saturated fat can be traced back to an epidemiological study conducted by Keys [11], commonly known as the Seven Countries Study. Keys [11] showed that the percentage of caloric intake from saturated fat was positively associated with coronary-related deaths, whereas a negative association was found from monounsaturated fat consumption [11]. Since this time, many concerns regarding the validity of the Seven Countries Study have been raised [12], as well as multiple observational and experimental trials have since shown contradictory results regarding the deleterious health effects of saturated fat consumption [7, 8, 13, 14]. Some studies have shown positive associations between the consumption of saturated fat and the risk of CVD [8, 14], whereas others have not [7, 13] a single study has shown an inverse association between the consumption of polyunsaturated fat and CVD [13], whereas again, several others have not [7, 14]. The aforementioned collection of studies on this matter indicates that there is no consensus in the literature regarding the consumption of saturated fat and an increased risk of CVD.

The major sources of dietary fat have changed within the past century [15], with the use of butter and lard being slowly replaced by inexpensive and more versatile plant-based oils and shortenings. With the introduction of mechanization, vegetable oils were becoming less expensive than animal fat sources. As a result, researchers had been putting forth tremendous effort toward changing the characteristics of fats, specifically changing the melting point of inexpensive oils to manufacture margarines and shortenings to act as lard and butter substitutes [6]. In certain cases, animal-derived shortenings, such as lard and compound lard (vegetable oil mixed with lard), became perceived as being unhealthy, unhygienic, and adulterated due to their association with the unsavory conditions in the meat-packing industry and popular marketing of all-vegetable shortenings [6].

A major technological breakthrough in the fats and oils industry was hydrogenation. Vegetable oils could be stabilized by hydrogenation and made more functional for inclusion into food products, by saturating double bonds and turning liquid oil into solid fat. Fully hydrogenated oils yielded brittle fats; however, partial hydrogenation created semi-solid, plastic fats that could be used for many food applications. Partial hydrogenation transforms *cis* unsaturated double bonds to *trans* double bonds to create triglycerides of higher melting points, which are solid, but plastic, at room temperature. This was the method of choice for transforming oils into fats, while still claiming high unsaturated fatty acid levels. This is achieved by varying the temperature, pressure, agitation, and catalyst concentration of the hydrogenation reaction, thereby creating a number of different hydrogenated fats with tailored characteristics in structure, texture, lubrication, tenderness, and aeration for the intended food product [6]. It was thought that since these hydrogenated fats came from healthful vegetable oils, that originally contained low saturated fat and cholesterol contents, they were a healthy alternative to animal-derived fats. Hydrogenated oils became ubiquitous in the food industry, with the replacement of butter and lard with margarines and shortenings in cakes, cookies, chips, breads, icings, fillings, etc. However, partially hydrogenated oils (PHOs) contain *trans* fats, which have now been identified to cause negative health effects [14, 16, 17]. It is important to note that *trans* fats also naturally occur in certain foods, such as meats and dairy products. These ruminant *trans* fats are present in very low amounts and have not been conclusively associated with the negative health effects of industrial-produced *trans* fats [18, 19].

In 2003, the US Food and Drug Administration (FDA) mandated that all packaged food manufacturers must label *trans* fat content on their products [20]. In 2007, Heath Canada stated that *trans* fats should be limited to 5% of the total fat content in the food product and that spreadable margarines should be limited to 2% of total fat content [21]. The Minister of Health called on the food industry to comply with these recommendations within 2 years and began a monitoring program to track the *trans* fat content in specific food products. In 2009, the World Health Organization (WHO) deemed *trans* fat produced by partial hydrogenation of fats and oils to be considered an industrial food additive that demonstrated no health benefits and a clear risk to human health, and recommended *trans* fat should consist of less than 1% of an individuals' daily caloric intake [22]. Due to the continuing mounting evidence confirming the negative health effects of *trans* fatty acid consumption, the generally recognized as safe (GRAS) status of PHOs was removed by the FDA with a 3-year compliance period [17]. Scientific evidence has shown that the consumption of *trans* fat from PHOs is associated with increased levels of low-density lipoprotein (LDL) cholesterol, decreased levels of high-density lipoprotein (HDL) cholesterol, and increased levels of plasma triglyceride. All three factors are well-defined risk markers for CVD and promote insulin resistance in humans, which leads to the onset of pre- and type 2 diabetes [14, 16, 17]. In 2017 Health Canada banned PHOs in foods completely (https://www.canada.ca/en/health-canada/services/food-nutrition/public-involvement-partnerships/modification-prohibiting-use-partially-hydrogenated-oils-in-foods.html), followed by the U.S.A. in 2018 (https://www.fda.gov/food/food-additives-petitions/final-determination-regarding-partially-hydrogenated-oils-removing-trans-fat). It is rather remarkable this took so long. Since the early 1990s [23], it has been known that *trans* fats increase LDL, triacylglycerols (TAGs), and insulin levels, and reduce beneficial HDL, all factors which increase the risk of developing CVD [24]. Only in 2006, 16 years after the deleterious effects of *trans* fatty acids on human health were conclusively shown, it became mandatory to claim *trans* fatty acids on food labels. The processed food industry remained heavily reliant on the partial hydrogenation of vegetable oils to produce solid fats until 2017–2018, and many continue under grandfather clauses. Now the focus has changed to saturated fat. The reduction of saturated fat in the diet has been a long-standing recommendation. The first dietary guidelines in the United States were published by the American Heart Association in 1957, and later by the US Senate in 1977. In regard to fat consumption, they recommended overall fat consumption should account for a total of 30–40% of total caloric intake with a reduction of saturated fat consumption to upwards of 10% of total caloric intake and balance the remainder with poly and monounsaturated fat sources [25]. Since Keys' early work in the 1950s, it has been known that increased SFA consumption is positively correlated with increased plasma total and LDL cholesterol levels, a marker of risk in CVD [26]. Replacing SFA with *cis*-PUFA or *cis*-MUFA is more favorable in terms of serum lipid levels than replacing carbohydrates with SFA [27]. For total and LDL cholesterol, the most favorable effects were seen in *cis*-PUFA, and overall health benefits have been correlated to increased consumption of mono and poly-unsaturated fatty acids which can be seen in Fig. 3.1.

Mensink [27] also found that increasing intake of steric acid did not appear to have a significant effect on these or other serum lipid values, concluding it does not negatively affect cardiovascular health, shown in Fig. 3.2.

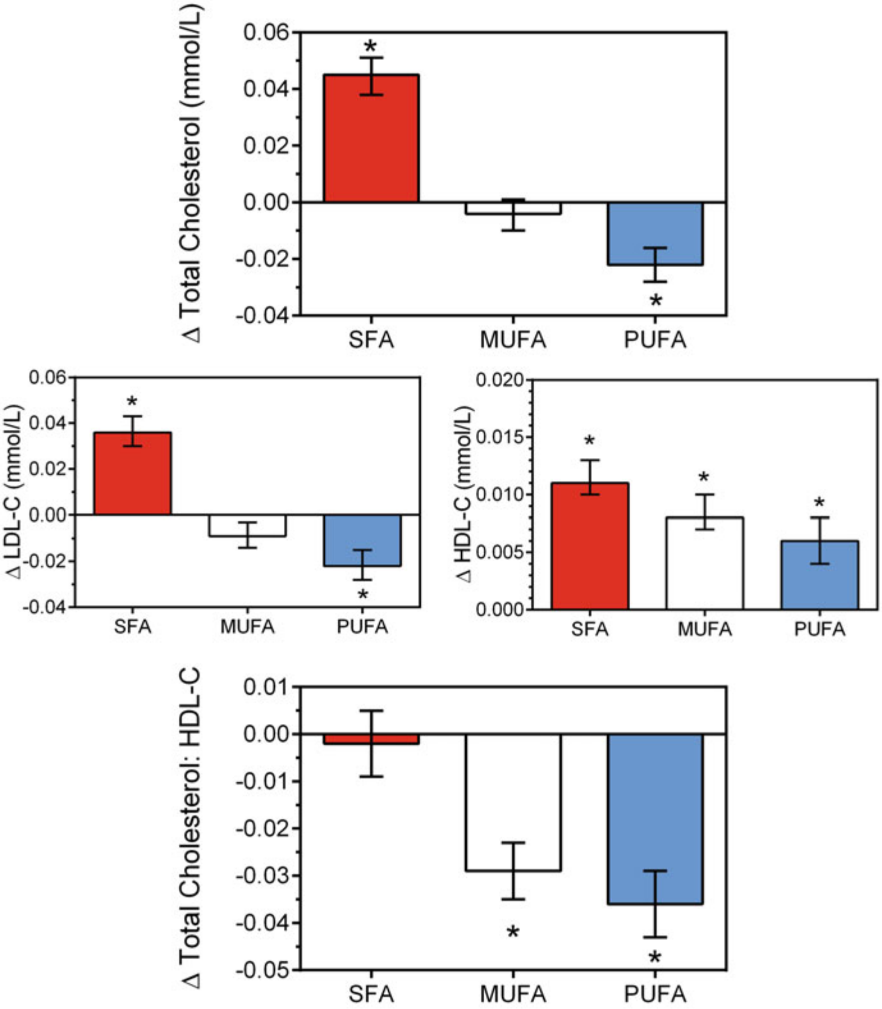

Fig. 3.1 Effect of saturated- (SFA), mono- (MUFA), and poly-unsaturated (PUFA) fat on serum lipoprotein levels, data from [27]. HDL: high density lipoprotein, LDL, low density lipoprotein

Based on the American Heart Association general guidance and recommendation, only 5–6% of total daily calories should be achieved by saturated fat. This means for a diet of 2000 calories a day only about 13 g of saturated fat per day is recommended (https://www.heart.org/en/healthy-living/healthy-eating/eat-smart/fats/saturated-fats).

Removing *trans* fat and PHOs from processed foods has already been undertaken by many companies; however, *trans* fat can still be abundantly found in products such as frozen packaged baked desserts, baked goods, coffee creamers/whiteners, spreads, fried foods, refrigerated doughs, vegetable shortening, etc. [21]. Removal of

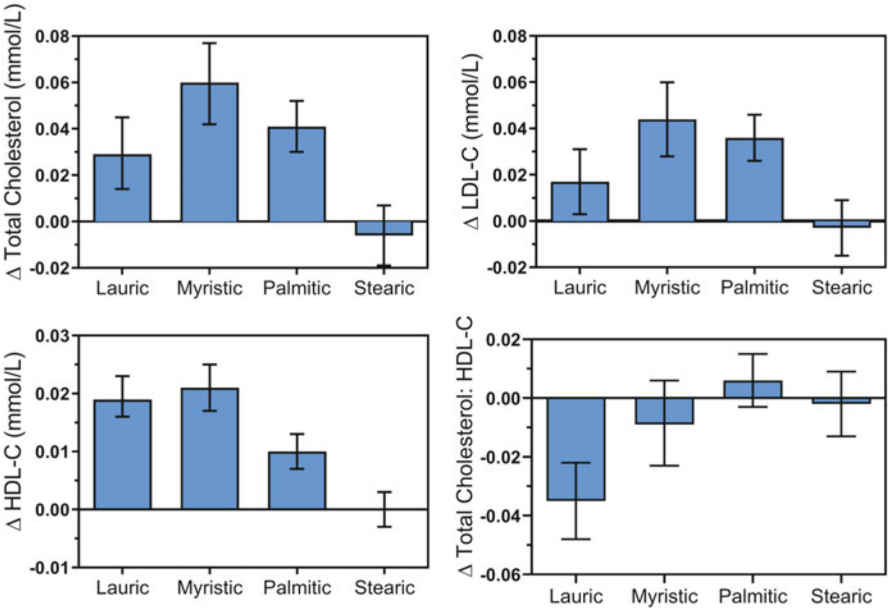

Fig. 3.2 Effect of consumption of saturated fatty acids on serum lipoprotein levels, data from [27]. HDL: high-density lipoprotein, LDL, low-density lipoprotein

saturated fats from animal sources has also been undertaken with the introduction of low-fat milk and meat products; however, much of the time, these saturated fats are replaced with carbohydrates. This strategy of saturated fat replacement, however, has been a cause of concern because low-fat, high-carbohydrate diets are associated with a different set of health issues including dyslipidemia, a component of metabolic syndrome [28–30].

Many food products are formulated with high amounts of saturated fat and/or *trans* fat because they contribute to the product's flavor, texture, mouthfeel, and functionality [31]. Research on how to replace these fats in food products without negatively affecting the organoleptic properties has been a topic of concern for the food industry and researchers alike. From an industrial perspective, a "drop-in replacement" or an ingredient that is completely interchangeable in a process is of the highest value. Ideally, changes to a product recipe should have minimal effects on product processing and the final properties of the product. Any changes to how a product must be processed upon the addition of the replacement ingredient can affect the quality of the final product and may incur losses, financial or other. Researchers must then be able to develop a suitable replacement that is both functionally and economically feasible. It is also important to consider other factors when seeking replacements for fats and oils, such as environmental sustainability, which is why the use of tropical oils, such as palm oil, is of concern [32]. In the last couple of years, a lot of attention has been drawn to the biodiversity loss in tropical rainforests due to the cultivation of both palm and coconut oil, predominantly in Malaysia and

Indonesia. These rainforests are biodiversity havens and act as massive carbon sinks. When these tropical forests are cut down and turned into palm oil plantations, a large increase in carbon emissions results. Conversely, palm is also the crop that has the highest yield per hectare and could be a more sustainable oil if produced responsibly [33]. Notable here are the recent Roundtable on Sustainable Palm Oil initiatives (https://rspo.org). Coconut oil is another prominent oil, whose production has expanded due to the demise of PHOs and the rise in the plant-based food market in which coconut oil is a main ingredient. Coconut oil is cultivated in more than 90 countries, but more than 80% of virgin or refined coconut oil is produced in Indonesia and the Philippines. Similar to the palm tree, the coconut tree grows in the same tropical forests with a high degree of biodiversity.

Besides environmental concerns about coconut oil and palm oil production, during these years, a big shift in food sources for the human diet has been observed, and one of the main routes to change this situation is the transition from animal-based foods to plant-based food products. Many scientists have been focused on mimicking the physicochemical, rheological, and textural properties of the food's target components (proteins, fats, and carbohydrates) and overcoming challenges introduced through the native organoleptic and palatability properties commonly associated with plant-based foods. However, as mentioned earlier both palm oil and coconut oil are not sustainable solutions for this change at the moment.

Recently, the use of edible organogels, or oleogels, has become a popular strategy to replace saturated fats in a variety of food products [31, 34–38]. Traditionally, fats are structured with TAGs, which provide a hierarchical crystalline structure that binds and traps oil that in turn provides structure and desirable functionality to numerous food products such as cheese, butter, and lard [1, 2, 39–41]. It is within these TAG structuring agents that *trans* and saturated fatty acids are present. Organogelation is a novel strategy that involves adding a gelator or a structuring agent to liquid oils, which subsequently forms a network that gels the oil and imparts a solid-like character that is analogous to fat [35, 42]. These networks differ from the structures arising from TAG crystallization, but have similar functionalities in numerous food applications, making them promising alternatives [43]. Oleogels can mimic animal fats and if necessary, minimize saturated fat content while obtaining the same functionality that saturated fats provide [35].

Many structuring agents have been identified that can be added or prepared with oil in order to mimic the functionality of hardstock fats that are both free of *trans* and have reduced saturated fat content. Direct approaches for obtaining organogels include structuring agents such as monoacylglycerols (MAGs) and diacylglycerols (DAGs), ricinelaidic acid, 12-hydoxystearic acid (12-HSA), ethyl-cellulose (EC), ceramides, and wax esters [34, 35]. Another recent method found is the use of indirect structuring agents, which include foam and emulsion templates made from modified polysaccharides and proteins to form functional structured oils [43].

One of the benefits of organogels is their high degree of versatility. For example, different oils, structuring agents, and mixtures of the two can be used to replace *trans* and saturated fats. These combinations result in structured oils with different functionalities, which then can be applied to different food products that have specific fat

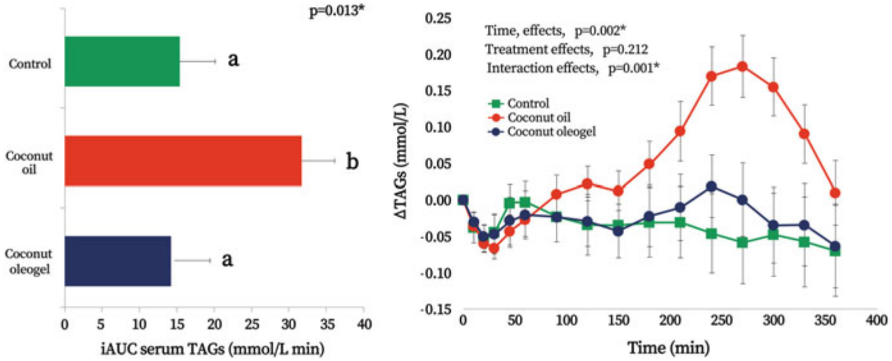

Fig. 3.3 Serum triacylglycerol changes following consumption of a carbohydrate-rich breakfast alone (control), with coconut oil, or with coconut oleogel. Bar graph corresponds to the area under the curve for each of the treatments. (Reproduced from [52] under the terms of the Creative Commons Attribution-NonCommercial 3.0 Unported Licence (CC BY-NC 3.0))

requirements. Multiple studies have been conducted in order to understand how these structured oils behave and affect the properties of different food applications including frankfurters [44], ice cream [45], chocolate [46, 47], cookies [48], and cream filling [49]. The understanding of food systems has become increasingly important in order to design its functional components, which in this case, is its fat content. As a result, further research needs to examine individual food systems in order to understand the effectiveness of replacing saturated and *trans* fat with structured oils. Furthermore, certain oleogels alter the physiological response of lipid metabolism (by decreasing rate of lipolysis) and can be used as an improved delivery system for lipid-soluble bioactive molecules and micronutrients [50, 51]. Oleogels open an exciting area of research whereby fats and oils that have demonstrated negative health effects can be structured in such a way to alter the metabolism of lipids, and minimize these effects [52]. For example, Tan et al. [52] demonstrated that ingesting oleogelated coconut oil (with EC) almost entirely eliminated the serum triglyceride peak after consuming a high-fat meal. These results were not seen in unstructured coconut oil (Fig. 3.3). Similar results have also been seen in structured emulsions [53, 54]. Rice bran wax (RBW) oleogels have also shown hypolipidemic and hypocholesterolemia affects post-consumption [55].

Figure 3.4 outlines the different mechanisms used to structure fat mimetics (oleogels). This section will review the most promising methods used to structure edible oils, their applications in food systems, and their limitations.

Many strategies of oil structuring have been reported using multiple different structuring agents that are food-grade, economically viable, and are able to impart desirable physical properties at low concentrations [31, 34, 35]. These structuring agents can be loosely defined under the following categories: crystalline particles, crystalline fibers, structured emulsions, polymeric strands [34, 35], and more recently inorganic particles [43]. The following section will provide a brief summary of the aforementioned novel oil structuring methods, while the section after will

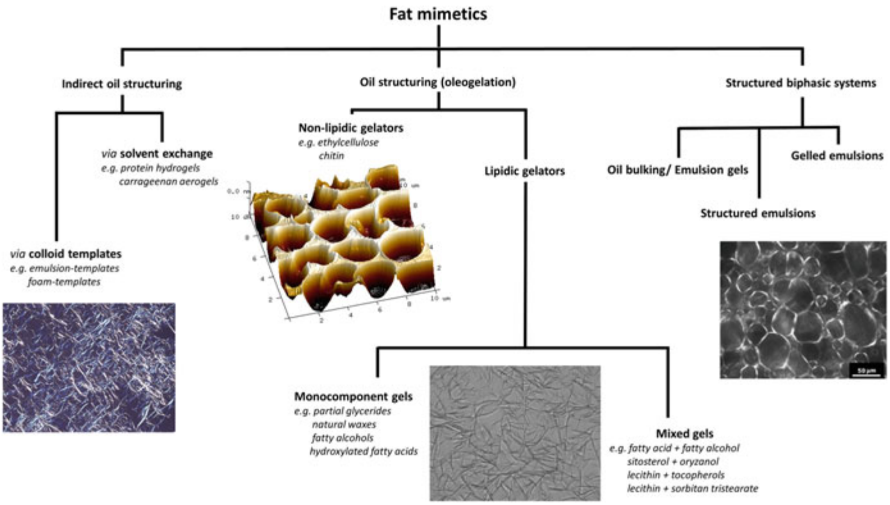

Fig. 3.4 Different fat mimetics strategies. (Reproduced from [42] with permission from Elsevier)

focus on more specific systems. In addition, novel direct structuring of oil into solid fat through enzymatic conversion of native TAGs to MAGs and DAGs and the potential of enzymatically modified systems as the oil phase of oleogels to produce adipose tissue mimetics will be discussed.

3.2 Oleogels and Oleogelation Strategies

3.2.1 Crystalline Particles

3.2.1.1 Partial Glycerides

Monoglycerides and diglycerides are a very promising class of organogelators as these molecules are natural components of edible oils and are already in common use in foods. MAGs consist of a single fatty acid esterified with a hydroxyl group attached to the end or middle carbon of the glycerol. Thus, MAGs are generally described as amphipathic molecules, having a polar “head” with no charged functional groups and a hydrophobic “tail” formed by the alkyl chain of the fatty acid. This gives them the ability to efficiently structure oil at concentrations of 10 wt% [56–58]. There are no restrictions for the use of MAGs in food products, making them an attractive structuring agent. Further, only a low concentration of MAG is needed to structure oils [56, 57]. MAGs are metabolized in a more favorable manner compared to TAGs leading to decreased body fat accumulation [59, 60].

MAGs are optically active molecules that exist as two chiral isomers, denoted by *D* and *L*. The application of MAGs in food systems is mostly associated with their emulsifying capacity, favoring the formation of emulsions and foams as well as preventing starch retrogradation in bakery products through the development of MAG–amylose complexes [35, 61]. However, an important property of MAGs in pure and commercial forms is their capacity of structuring vegetable oils and oil in water emulsions, through the formation of several liquid crystalline structures [61]. MAGs have higher crystallization and melting temperatures (both saturated and unsaturated) compared to triglycerides (TAGs) [62]. Therefore, the use of MAGs as organogelators has merited the attention of many research groups to produce fat mimetics. MAG-based oleogels are prepared by direct dispersion and melting of a relatively low amount of MAG in oil followed by cooling, resulting in a complex solid network of crystalline particles that can entrap the oil. Although a 2–3 wt% of saturated MAGs in oil has been reported as the critical gelling concentration, greater amounts (5–10%) are required to obtain rheological and textural properties comparable to those of commercial plastic fats [63]. When using MAGs, it is also possible to incorporate large amounts of water, which in some food applications comes as a great advantage, particularly in biphasic systems containing both oil and water phases [35, 64]. The crystallization and polymorphic properties of MAG crystals are very important in determining their functional properties in food products. Although at least five polymorphic forms can be found for triglycerides, MAGs crystalize in α, sub-α, and thermodynamically stable β forms. During cooling, the MAGs form an inverse lamellar phase (at a temperature above the Krafft or chain melting temperature) with a hexagonal packing similar to the α-gel phase in the aqueous systems. Further cooling of the system (below the crystallization point of MAGs) results in the formation of the sub-α phase with an orthorhombic crystalline packing. Both phases are metastable and change into a triclinic subcell packing with a higher melting point (β polymorph) with time [35]. Processing parameters such as MAG composition (saturated/unsaturated and chain length) and concentration, heating and cooling temperatures, agitation, shear rate during cooling, and storage time affect the kinetics of crystallization and the size of MAG crystals [63]. These changes in microstructure translate into the macroscopic mechanical features as evident in the rheological behavior of MAG oleogels [65, 66].

Most of the research about MAGs as gelators is based on MAGs of palmitic and stearic acids. Monostearin and monopalmitin have been successfully applied to gel a variety of oils including high oleic sunflower oil (HOSO) [66–73], rapeseed oil [74], coconut oil [72], flaxseed oil [75], canola oil [76, 77], safflower oil [78], hazelnut oil [79, 80], olive oil [56, 57], corn oil [81], and cod liver oil [82]. However, very recent studies have reported the use of MAGs of shorter-chain fatty acids such as caprylic, capric, and laureate acids [71, 74, 83] and longer-chain fatty acids such as behenic acid [71, 74]. Ferro et al. [72] and Valoppi et al. [84] studied the effect of oil type on the structure and physical properties of MAG-based oleogels. Stronger gel networks were formed with the increase in oil viscosity and reduction of oil polarity [84]. Furthermore, the degree of saturation of oils affected oleogel properties. Long-chain monounsaturated fatty acids favored the packing of MAG crystals in a cohesive gel [72].

The main concern related to using MAGs as structuring agents is their lack of structural stability. MAG structural destabilization is due to a progressive rearrangement and separation of chiral MAGs (*D*- and *L*-isomers) [63] and a tendency to transform from the α to β-polymorph form over storage time whereby hardness and brittleness increase, while oil binding capacity is lost [85], causing a gritty texture and oiling off [43]. However, their stability can be enhanced by the addition of other co-emulsifiers and changes in processing conditions. Polyglycerol fatty acid esters (PGE), sodium stearyl lactylate (SSL), and propylene glycol monostearate (PGMS) are all examples of surfactants that help stabilize the α-gel state and can be used to enhance stability in food products including low-fat shortenings, and aeration properties of cake batter [86]. Anionic, mono-valent, and small head structures are all characteristics of coemulsifiers that enhance the stability of MAG-structured oils. Recent investigations have also indicated improved rheological properties and physical stability of oleogels based on a mixture of MAGs and phytosterols [87–89]. For example, a 3:1 monoglyceride:phytosterol in sunflower oil can replace 50% of pork fat in Frankfurt sausages ending in a product with strong gel strength, and similar elasticity, cohesiveness, and oxidation levels [88]. Phytosterols can prevent the aggregation of MAGs into larger structures. This results in a system with plenty of small crystals, which typically produce a harder gel [87]. Moreover, oleogels formulated with MAGs and waxes from different sources [75, 90, 91], hydrogenated fat [92], lecithin [93], a mixture of wax and EC [94] and EC [78] have also been studied to evaluate their synergistic properties and ability to limit the sub-α to β polymorphic transition of MAGs. Palla et al. [95] attempted to match the texture profile and oil-binding capacity of a MAG-oleogel with that of a commercial margarine by studying the effect of preparation variables (MAG concentration, heating temperature, stirring speed, and cooling temperature) on oleogel characteristics. MAG concentration and cooling temperature were found to be the most contributing factors to oil binding capacity, and textural and rheological parameters [95]. In addition, new technologies such as high-intensity ultrasonic have been found to improve the physical properties of MAG-based oleogels [68, 69]. Shear and cooling rates can be used to alter the structure of the crystalline network and ultimately influence their physiochemical properties in food [57, 96]. When using MAGs, it is possible to incorporate large amounts of water, which in some food applications comes as a great advantage, particularly in biphasic systems containing both an oil and a water phase [58, 97]. Their strong emulsifying ability makes them excellent candidates for cream-like products [65]. Another partial glyceride-stabilized oleogel strategy exploits the use of DAGs. Similar to MAGs, DAGs have high melting points and desirable health benefits, making them an interesting candidate for oil structuring. DAGs are diesters of fatty acids with glycerin and can exist in two isomers: 1,2- (or 2,3) DAGs and 1,3-DAGs. While 1,2-DAGs have a melting temperature similar to that of TAGs, 1,3 isomers show a melting temperature that is approximately 10 °C higher than TAGs of the same fatty acid composition. In addition, due to different metabolic pathways, 1,3-DAGs inhibit body fat accumulation and reduce postprandial TAG levels in human serum [62]. The crystallization behavior of 1,3-DAGs and 1,2-DAGs differs significantly. 1,3-DAGs crystallize in β configuration either a high-melting polymorphic form β_1, or a lower-melting polymorphic form β_2 [98]. Although β crystals are related to oil loss and sandiness in fat products, they are not necessarily an issue if small β crystals are formed. In fact, small β crystals are also capable of forming a stable crystal network structure with desired rheological properties [98]. Xu et al. [99] reported that fat blends containing DAGs had a stronger network structure with a more solid-like nature compared to the fats structured purely with TAGs. These merits suggest the promising use of DAGs in the fabrication of oleogels. Tavernier et al. [98] investigated the effect of MAGs (1, 2, 4 wt%) on the crystallization behavior of 20 wt% DAG sunflower oil oleogel. MAGs delayed the onset of nucleation and retarded the crystal network development by 1,3-DAGs. The effect of MAG-DAG interactions on the microstructure and physicochemical properties of the oleogels was further explored by Wang et al. [100]. In this study, the minimum concentration for DAG to form a solid-like gel was found to be 8 wt%. Different MAG/DAG weight ratios (0:10, 1:9, 3:7,5: 5, 7:3, 9:1, and 10:0) were then applied to prepare mixed oleogels with total gelator concentrations of 8 and 12 wt%. MAG-based oleogels produced significantly higher hardness, possibly due to the higher crystallization rate, higher melting point, and higher oil binding capacity of MAG molecules. The mixture showed reduced crystallization enthalpies compared to mono-component oleogels, indicating eutectic behavior and co-crystallization of MAG and DAG molecules. However, the oleogels with higher MAG ratios showed lower solid fat content at low temperatures. Oleogels with good plasticity and crystal morphology were obtained using MAG: DAG ratio greater than 5:5 [100].

The interfacial activity and strong network structure formed by DAG have also led to research on DAGs as a structuring agent in emulsions or foams by forming Pickering particles. Oleofoams can be fabricated by whipping the suitable oil phase and have shown great potential as low-calorie aeration systems. However, saturated lipids or a large amount of surfactants are commonly required [101]. In work conducted by Lei et al. [102], oleogel prepared with 10 wt% long-chain DAG was found to be a feasible substitute for hydrogenated palm oil (HPO), showing comparable rheological and thermos-sensitive properties. The incorporation of 2 wt% HPO into the system contributed to ultra-stable oil foams with higher viscoelasticity and smaller bubble size. To provide desirable oil structuring and whipping ability, Qiu et al. [101] suggested the combination of DAG and β sitosterol as gelators. The gelators interacted through hydrogen bonding, which had a synergistic effect on the formation of a stable physical barrier and the prevention of air bubble destabilization. In subsequent work, the authors compared the performance of pure diglycerol laurate in an oleogel and oleofoam with lauric acid and its other glycerides (glycerol monolaurate and triglyceride laurate). Lipid crystal size and polymorphism largely affected the whipping ability and foam stability. DAG crystals exhibited smaller size and β and β' polymorphs with uniform distribution at the bubble surface, which contributed to the formation of ultra-stable oleofoams. The lauric acid stabilized system had a comparable over-run to that of DAG oleofoams while the MAG and TAG mixtures showed lower over-run. According to the results, a variety of plant oil foams with high stability (up to 8 months) can be stabilized by DAG crystals [103].

In conclusion, considering the natural origin of MAGs and DAGs and their unique physicochemical and physiological functions, partial glyceride-based structured fats can be designed not only for solid fat replacement purposes but also to modulate lipid digestion. These clean-label multifunctional systems have been used in a wide range of food products with promising results. Future work in the partial glyceride-oleogel field requires a focus on technological and health-promoting applications as well as scaling up laboratory experiments to pilot and industrial scale.

3.2.1.2 Waxes

Plant-based waxes are also capable of structuring oil at low concentrations (1–4 wt%) by forming crystalline particles that aggregate and form a network that entraps oil [34, 35]. Some major sources of wax that have been studied include RBW [104], sunflower wax (SFW) [105], candelilla wax (CLW) [106], carnauba wax (CRW) [107], shellac wax [108], beeswax (BW) [109], and sugarcane wax [107]. Natural waxes are promising gelling agents for vegetable oils because of their strong oil-binding capacity, economic feasibility, and wide availability to be used in the food industry [42, 106, 110]. Chemically, waxes are composed of mainly long-chain esters from fatty acids and fatty alcohols along with minor components such as n-alkanes and sterol esters [104, 111, 112]. Low concentration of wax (1–4%) is needed in an oleogel formulation to obtain gelling properties [104, 113]. This makes them economically viable along with the fact that waxes are often by-products of oil refining [113]. Table 3.1 shows the most promising waxes, their chemical composition, and their melting temperature. All of the following waxes included in Table 3.1 have gained GRAS status from the FDA, excluding SFW.

There is a strong relationship between wax composition and crystal morphology. Smaller crystals form upon faster cooling and increased shear rates. Wax esters crystalize in mono-molecular chain packing with alkyl chains either in a tilted or orthogonal orientation [115]. Melting points and molar heats of fusion are related to the position of the ester bonds, the more asymmetrical the bonds the lower the melting points and molar heats. Crystals that are uniform in size and range between 1 and 5 μm produce a smoother texture and soft mouth feel [116, 117]. Waxes tend

Table 3.1 Chemical composition and melting temperature of vegetable waxes and beeswax [109, 113, 114]

	Esters (%)	Free fatty acids (%)	Free fatty alcohol (%)	Hydrocarbons (%)	Resins (%)	Melting temperature (T_m)
RBW	92–97	0–2	–	–	3–8	78–82
SFW	97–100	0–1	–	–	0–3	74–77
CLW	27–35	7–10	10–15	50–65	–	60–73
CRW	84–85	3–3.5	2–3	2.5–3	6.5–10	80–85
BW	71	12	–	14	6	63.15

BW beeswax, *CLW* candelilla wax, *CRW* carnauba wax, *RBW* rice bran wax, *SFW* sunflower wax to leave a waxy taste in the mouth and can be gritty, therefore, tailoring the processing parameters to make the wax crystal structure optimal is essential to limit undesirable sensory properties. Synergistic mixtures of waxes have also been reported. Ghazani et al. [118]. discovered synergism between SFW and RBW, as well as CLW and BW, resulting in increased hardness, elastic constant, plasticity, and oil binding capacity of oleogels compared to each counterpart on their own. These mixtures with olive oil demonstrated similar physical characteristics to commercial margarines. Wax oleogels have also had many successful applications in the bakery sector, replacing margarine and spread products with minimal sensory differences [48, 119, 120]. Wax oleogels have also been used to lower saturated fats in deep fat frying mediums [121].

Although wax oleogels are great structuring agents, they do come with some limitations, namely, sustainability issues with the extraction of certain waxes, such as CLW [122], changes in rheological properties in systems over time (stability), cloudy appearance, and more importantly, waxy mouthfeel in food products. Further questions also exist regarding digestibility and health risks/benefits associated with long-term consumption of these waxes [123].

3.2.1.3 Fatty Acids and Fatty Alcohols

Fatty acids and fatty alcohols were some of the first molecules studied to structure oils. These low-molecular-weight compounds are able to form supramolecular structures through non-covalent interactions that structure oil very effectively at low concentrations (~1% w/w) [124–126]. An attractive attribute of these systems is their similarity of crystallization behavior to TAGs [127]. To make fatty acid /fatty alcohol gels, the mixture is simply heated, and gelation occurs upon cooling. Stearyl alcohol /stearic acid (SOSA) has the highest oil structuring capacity in edible oils compared to any other fatty acid/fatty alcohol mixture [125]. Gandolfo et al. [125] demonstrated that a specific mixture of fatty acids and fatty alcohols of similar chain lengths displays a synergistic effect in terms of oleogel strength. The 7:3 (w/w) SOSA mixture formed smaller crystals with needle-like morphology than the other proportions. These small crystals had a greater surface area, higher oil binding, and formed a stronger network [125, 128]. Blach et al. [124] revisited this system and demonstrated that the desirable synergistic effect was due to an increase in the spatial distribution of network crystalline mass. Both studies used a 5% structurant concentration, and the same effect was observed using sunflower oil [125, 128]. Rapeseed oil's optimal ratio was slightly different than for soybean and sunflower oil, with an optimal ratio of 8:2 w/w [125]. These optimal ratios displayed no oil loss upon centrifugation and had great stability, making them excellent candidates for food applications [124]. The lowest concentration of these molecules that can form a crystal network was 3.5% [128].

Food applications for SOSA include margarine, sausages, and Dutch croquettes [128]. SOSA can also be used in combination with other structuring agents (such as EC), which results in enhanced plastic flow behavior to resemble commercial margarine [126]. Lecithin has also been shown to display synergism when mixed with various fatty acids and/or fatty alcohols [127]. Lecithin and tocopherol [129], β-sitosterol and oryzanol [130], and lecithin-stearic acid-water [131] provide flexibility to alter desirable properties for specific commercial applications at low gel concentrations [132].

3.2.2 Crystalline Fibers

Oil structuring can be achieved by self-assembled fibrillar networks (SAFIN) of low molecular weight gelators (LMOG), for example, 12-HSA, ricinelaidic acid, and phytosterols with oryzanol [35]. These gelators are capable of forming helical and twisted crystalline ribbons, hundreds of micrometers long, which immobilize oil.

3.2.3 Structured Emulsions

One of the first oil gels that demonstrated viability for use in food applications were high-internal phase emulsion (HIPE) gels, consisting of oil encapsulated in multilamellar crystalline MAG vesicles [58, 133, 134]. The properties of MAG gels were exploited to create a simple and highly stable oil-in-water emulsion gel with 27–70% (v/v) water, 66–27% (v/v) oil, zero *trans*, and low saturated fat content [58]. The resultant structured emulsion exhibits a fat-like appearance with highly versatile shortening functionality and is commercially available today as AAK's AkoBiscGO (https://www.aak.com/applications/bakery/biscuits-and-cookies/AkoBiscGO/), trademarked under the name Coasun™ (https://www.coasun.com/). The addition of a large percentage of water to make CoaSun™ can significantly cut production costs [44].

The gel preparation involves dispersing of hot oil-monoglyceride-charged co-surfactant mixture in hot water by vigorous mixing followed by cooling, resulting in effective microencapsulation of oil within crystalline shells of monoglyceride and a solid-like structure.

Once emulsified, a monoglyceride monolayer surrounds the oil droplets and self-assembles to form a highly hydrated, multilamellar La state at temperatures slightly above the Krafft temperature of the monoglycerides. Upon cooling, the fatty acid chains of the monoglycerides crystallize in a hexagonal conformation (a subcell polymorph), referred to as an alpha gel, with high amounts of water entrapped between stacked bilayers. This, so called, a-gel state dehydrates and eventually transforms into an anhydrous crystalline phase (b' or b polymorph), referred to as the coagel or β-gel [58]. Although MAGs are incorporated at low levels (a few percent), yet they are able to stabilize the emulsion. A co-surfactant enhances the formation and stabilization of the a-gel state and adjusts the vesicle surface charge, affecting vesicle–vesicle interactions and rheological behavior of the emulsion

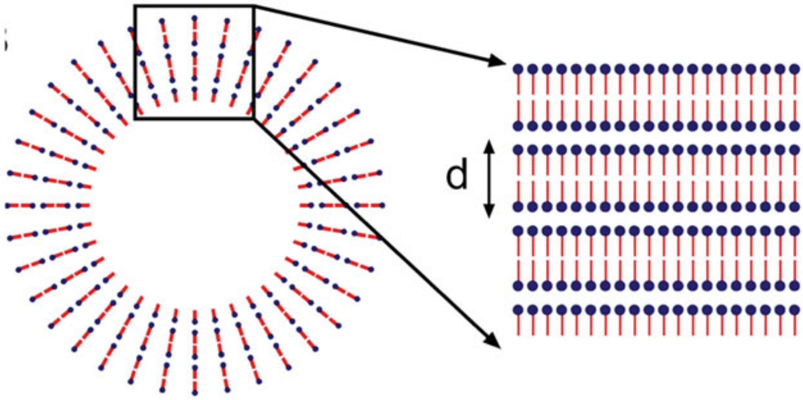

Fig. 3.5 Schematic of monoglyceride bilayers surrounding the oil droplets. Each of multiple bilayers could hold a layer of water between the hydrophilic head groups

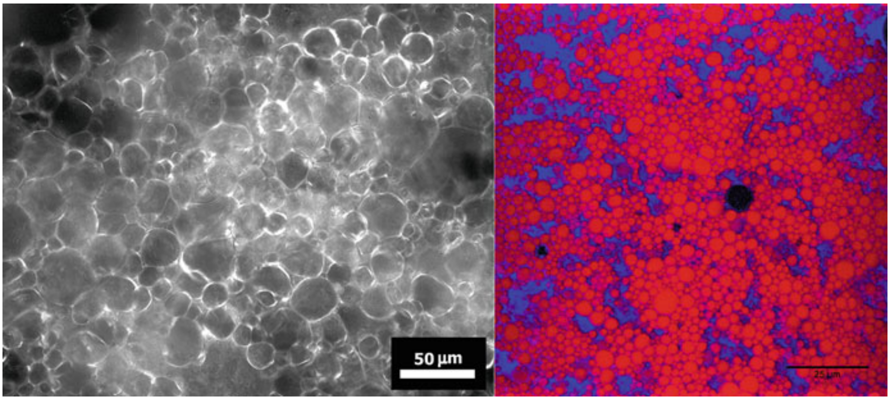

Fig. 3.6 Polarized light (left) and confocal microscopy (right) images of a structured oil-in-water emulsion (MAG gel)

[58]. Thus, the surface charge can be adjusted to convert a simple oil-water mixture into a fat-like material with desirable functional properties [58]. Figure 3.5 depicts a schematic of the outer walls of the droplet composed of stacked crystalline monoglyceride and water bilayers surrounding the oil. These droplets can be easily observed when using polarized light or cryo-scanning electron microscopy, as shown in Fig. 3.6.

Having very low SFA and zero *trans* fat content is one of the greatest advantages of these materials, making it possible to manufacture healthier food products, when prepared with canola oil (55.25%) and 40% water, CoaSun™ will contain 7–8% total saturates and can usually replace traditional shortenings on a 1:1 basis [135]. Furthermore, the unique structure of Coasun™ has shown the ability to regulate blood lipid and insulin response in humans. There is also a potential to incorporate nutraceuticals such as phytosterols, omega-3 fatty acids, and fat-soluble antioxidants into the water or oil phase of these structured emulsions, which increases their nutritional benefits [44].

Another method of creating structured emulsions was achieved by forming an organogel using shellac [136] or BW [109] in oil, and incorporating water into the molten organogel continuous phase in either the presence or absence of varying amounts of emulsifiers (e.g., Tween20, Tween80) and hydrocolloids (e.g., xanthan gum, XG). The molten organogel is subsequently cooled to entrap water droplets in the resultant crystalline organogel network. The shellac emulsions, which were prepared in the absence of any emulsifiers, consisted of 20 wt% water with either 1.6, 3.2, or 4.8 wt% shellac. This emulsion is stabilized by a combination of physical entrapment and steric stabilization as shellac both imparts structure to the continuous oil phase and accumulates at the water–oil interface, which prevents coalescence and phase separation [136]. The BW structured emulsions contained 3.75–4.50 wt% BW in the final emulsion and required the presence of emulsifiers and XG to remain stable [109]. It was speculated that Tween 20 or Tween 80 incorporation increased the hydrophilic-lipophilic balance (HLB) value, making the mixture more suitable for a water-in-oil emulsion. The addition of XG increased the viscosity of the water phase and decreased the sample melting point [109].

Alternatively, various food-grade hydrocolloids have been used to create oil continuous HIPE gels [137]. To develop such systems, the hydrocolloid solution is finely dispersed in an oil continuous phase at >70 °C and subsequently cooled to produce a high phase volume of closely packed gelled droplets that physically entrap oil and exhibit gel-like behavior [137]. The characteristics of used hydrocolloids affect the gelling efficiency of the internal water droplet phase and usually a combination of hydrocolloids is preferred. For example, galactomannans are known to show synergistic interactions with carrageenan, XG, and locust bean gum, leading to the formation of gel at low polymer concentrations [137].

3.2.4 Inorganic Particles

Hydrophilic fumed silica was able to structure sunflower oil to produce viscoelastic gels at concentrations of 10 and 15 wt% when silica particles were dispersed uniformly in oil under high shear [138]. At lower concentrations (2.5 and 5 wt%), the samples were viscous sols with negligible yield stresses. This was attributed to the highly concentrated samples having a higher degree of particle–particle interactions leading to a stronger fractal-like network formation which entrapped oil. In addition, a weak hydrogel containing locust bean gum and carrageenan could be mixed with the 15 wt% silica organogel to produce a bigel, which showed a higher gel strength than either component in isolation [138].

3.2.5 Hydrophobic Polymers/Oleocolloids

The only known direct polymeric organogelator and one that has shown great potential for use in food applications is EC. EC, a derivative of cellulose, is a non-water soluble, non-toxic, linear polysaccharide that is used as a thickener in foods. EC is made through a chemical modification process whereby cellulose is introduced to an alkaline environment and then exposed to chloroethane gas [139]. This induces the ethylation of the hydroxyl groups, which are exposed on the backbone of cellulose. The degree of ethylation is dictated by % ethoxy content which for most commercial food grades of EC lies between 48% and 49.5% (the range in which EC is soluble in organic solvents) [140]. Substituting the hydroxyl groups with ethoxy groups renders EC soluble in oil above its glass transition temperature ($T_g \sim 135$ °C) and upon cooling forms a three-dimensional entangled polymer network and a resultant gel. The functionality, microstructure, and processing conditions of these gels have been extensively studied [141–143]. Other food-grade polymers have been used to structure oils via an indirect methodology involving creating polymeric templated scaffoldings which are also capable of physically entrapping oil. EC oleogels are good substitutes to replace solid fats because of their similar fat globule size and microstructure [44, 144]. The strong mechanical strength of EC oleogels allows the gels to decrease the rate of lipolysis and serum TG postprandially compared to regular oils [51].

As mentioned, the process for preparing organogels using EC involves heating the oil-polymer mixture above its T_g, approximately 135 °C, and mixing it until the EC is dissolved (Gravelle and Marangoni, 2018). It is important to consider that the temperature of the mixture does not exceed 185 °C as there is a melting phase transition around this point and decomposition of the polymer starts. Upon cooling, EC will return to its rigid state creating gaps within the polymer network that physically entrap the liquid oil phase [145]. Various factors that affect the strength of the oleogel include temperature, cooling rate, the presence of surfactants, the type of liquid oil, the molecular weight of EC being used, and holding time during preparation. An increased holding time at T_g results in a stronger gel [146]. Because a high temperature is needed to create these gels, heat-induced oxidative degradation can occur, decreasing the shelf-life of the oil [147]. The molecular weight of EC also influences final gel strength, cross-over behavior, and gel point temperature [148]. EC is sold based on its molecular weight such that different molecular weights are tailored to different applications [149].

Many food applications have been studied including replacing some animal fat with EC oleogels in breakfast sausage [150], liver pate [151], salami [152], and frankfurters [143, 147]. It should be emphasized that in most of these applications only part of the animal fats was replaced in order to maintain consumer acceptability. EC oleogels have also been used to make heat-resistant chocolate [46]. EC can also be mixed with different molecules to manipulate the desired flow behavior and demonstrate synergism with respect to enhanced crystallization structure and increased plasticity. Gravelle et al. [126] demonstrated how the brittle behavior of

SOSA oleogels can be plasticized by the addition of EC allowing for plasticized behavior under deformation, mimicking a commercial margarine. Barbut et al. [153] also demonstrated that adding 1.5% or 3% sorbitan monostearate (SMS) to an 8% EC oleogel showed enhanced similarities in the hardness and color of beef fat in frankfurter sausages. In the USA, EC has obtained a "generally regarded as safe" (GRAS) status from the US Food and Drug Administration (FDA) for indirect food usage (21 Code of Federal Regulations section: 182.90) for use up to 5% (w/w) EC in oil. Depending on the type of oil used, the required amount of EC needed to form a gel differs. For canola and soybean oil a minimum of 6 and 4% w/w respectively are needed [143]. In the future, further research is needed on the consumer acceptability and sensory analysis of food products made with EC [154]. Another limitation of EC is it is not a natural ingredient. This stops many food companies from using it as many consumers steer away from long chemical names on food labels, seen frequently in plant-based products. The trend toward clean-label products has drastically increased in the last decade [155]. Ideally, a natural polymer could be found in the future with similar properties to EC.

3.2.6 Hydrocolloid-Stabilized Oil-Filled Gels, Foams, and Powders

Hydrocolloids are attractive agents to structure oils due to the lack of regulatory restrictions and because of their high molecular weight allowing them to form gel networks at low concentrations [42]. The majority of these hydrocolloids are proteins and modified polysaccharides and are thus hydrophilic and unable to be dispersed in oil like EC. Therefore, these polymers are added through indirect methods including emulsion and foam templates, which will be discussed. Finding proteins to use as structurants in oils has become an expanding area of research in the last couple of years. Proteins possess some key advantages including their price compared to other structurants, nutritional composition, clean label, and eligibility to be used in vegetarian and vegan products which are on the rise [156].

3.2.6.1 Emulsion-Templated Oleogels

Emulsion-templated oleogels are obtained by first creating an emulsion in which the aqueous phase (containing the hydrophilic polymer, a polysaccharide, or protein) is the continuous phase. The hydrocolloid stabilizes the oil-water interface and can get crosslinked to provide elasticity to the interface [157]. The continuous phase is then removed through oven drying, spray drying, or freeze drying, followed by shearing, resulting in a semi-solid material with a high concentration of lipid globules coated in the hydrocolloid creating the emulsion template [158, 159]. Gels structured through freeze drying resulted in harder gels when compared to oven-dried gels

[160, 161]. The structure and characteristics of the emulsion are closely related to the final properties of the oleogel [42]. Therefore, the structure of the oleogel is highly influenced by the functionality of the protein or polysaccharide. To use proteins in this structuring method they must be surface active and have the ability to coagulate [156]. Thankfully protein functionality can be modified through various processing parameters, more research needs to be done in this area to understand what is the structure of an optimal protein or polysaccharide for this application. Soy protein isolate thus far has been the most promising plant-based protein to form these gels in terms of its rheological properties [162]. Many protein oleogels tend to be much softer than the counterparts they are aiming to replace. As more research is done in this area and more proteins are explored in the upcoming years, they could make an excellent candidate for fat replacement with economic, environmental, and nutritional advantages.

In terms of polysaccharides, XG was used as a thickening agent and either hydroxypropyl methylcellulose (HPMC) or methylcellulose (MC) was used as emulsifying agents [158, 159]. Emulsions prepared with HPMC or MC alone resulted in coalescence and oil leaking upon drying. XG used in isolation could not emulsify, as it is nonsurface active, whereas, when incorporated in combination with HPMC/MC a more uniform oil droplet size distribution and stable dried emulsion was achieved [158]. In another study, it was found that the XG could also be used in combination with gelatin (a protein) to produce similarly structured oils where an increase of polymer concentration at the oil–water interface resulted in increased gel strength [159]. The microstructure of these structured oils shows distinct tightly packed oil droplets resembling that of a HIPE [158].

3.2.6.2 Foam-Templated Oleogels

The basic steps for obtaining a foam-templated oleogel are similar to those for emulsion-templated oleogels. To make a foam-templated gel, protein or polysaccharide is first mixed into an aqueous solution. Air is then introduced into the system (i.e., through homogenization) to create an aqueous foam. The aqueous foam is freeze-dried, becoming a "cryogel." Oil is introduced into the cryogel transforming it into an oleogel. Foam-templated gels tend to be softer compared to HIPE gels [108]. Advantage of this approach of forming oleogels is the environmental aspects of the processing steps, no high temperatures, harsh chemicals, or additives are used [163].

Patel and coworkers, the pioneers in this area, originally incorporated oil into a HPMC scaffold, [163]. In this work, an aqueous solution of HPMC was foamed mechanically to incorporate air and freeze-dried to create a porous structure that is able to adsorb a high amount of oil; however, since freeze-drying creates an open-celled structure, oil will be released under compression. To prevent this, the oil-saturated foams were sheared to disperse the polymer sheets and trap the oil [163]. This templating concept has also been used previously to partially (60%) and fully substitute pork backfat content in fermented sausages by adsorbing extra-virgin olive oil to a whey protein-based crumb and white pan bread [164]. Alessandro et al. [164] found no significant difference in weight loss of the oil-soaked whey protein-based crumb, white pan bread, and full-fat control products. In addition, taste, color, and odor characteristics of the 60% replacement with oil-soaked whey protein crumb were comparable to the commercial product; however, long-term stability was not examined. Another application for this templating concept is the use of cellulose-based freeze-dried foams as oil absorbents for cleaning oil spills in water [165]. The freeze-dried foams used for oil spill applications, typically are not food grade, which makes HPMC foams unique and useful for food applications. HPMC and MC have been used in the food, cosmetic, and pharmaceutical industries for decades and both have been granted a GRAS status by the US FDA. HPMC and MC are well known for their ability to create gels in aqueous solutions, but the literature is limited on their use in oil-based systems.

The functional properties of the final oleogel are directly related to the cryogel structure [166]. These gels can be used to decrease the density and caloric effects of food products [167]. Further, they can be tailored to change the texture, mouthfeel, appearance, and rheological properties of food products. Scaling up and stability of these foams need to be further researched [168]; however, they provide great potential for the replacement of *trans* and if desirable saturated fatty acids in edible applications.

3.3 Enzymatic Glycerolysis

Enzymatic glycerolysis (GL) is another technological alternative for obtaining zero trans, low saturated plastic fats, and improving the sustainability of the fat sources. The process converts oil-native TAGs into partial glycerides in the presence of glycerol and lipase, exploiting their higher crystallization and melting temperatures to directly structure the liquid oil [169, 170]. This is beneficial to consumers as partial glycerides are natural components of fats and oils and there is no change in the fatty acid composition (i.e., no increase in *trans* or SFA) of the lipid system. Therefore, readily available plant oils rich in unsaturated fatty acids can be modified to replace palm oil and other high-saturated-fat [169]. In addition to lower SFA intake, glycerolysis products (GPs) are of great interest due to the health benefits of DAGs on lipid metabolism such as inhibiting body fat accumulation and reducing serum postprandial triglyceride and LDL level, as stated earlier [62, 171]. Therefore, an edible unsaturated oil structured using GL would provide the nutritional benefits of MUFAs, PUFAs, and DAGs, while exhibiting the required solidity [169]. Another interesting property of GPs is their self-dispersion and emulsifying properties, showing their promise for use in emulsified products [169, 172] and as bioavailable lipid carriers in the formulation of functional foods and nutritional supplements [173].

Traditionally, GL is applied for simply producing partial glycerides for use as emulsifiers and other food ingredients. However, Marangoni group investigated the effectiveness of lipase-catalyzed GL for the novel purpose of fat structuring, where partial glycerides are not extracted after production [169]. This study was performed using cottonseed oil and non-specific *Candida antarctica* lipase B (2% w/w relative to the oil) at 65 °C for different time lengths and with varying glycerol to oil molar ratios (0.10:1, 0.25:1, 0.50:1, 1:1, 2:1, 4:1). A rapid increase of MAGs and DAGs content was observed during the first 18 h of reaction, which reached almost a plateau after 48 h. DAG content was maximized at 0.5:1 glycerol:oil ratio and decreased slightly at higher glycerol content while the 1:1 ratio resulted in the highest MAG production. Reaction conditions also had an impact on MAG species. The longer reaction time and the higher glycerol content increased monoolein and monopalmitin while having a limited effect on monolinolein content. Overall, results showed dramatic changes in solid fat content (SFC), crystallization behavior, microstructure, and oil binding properties of the oil followed by GL. At 5 °C, SFC increased from almost 8% in native oil to 29% in optimally structured cottonseed oil GP, with 21.7% solids remaining at 20 °C. Microstructure and network properties obtained at 1:1 glycerol:oil ratio resulted in high oil binding capabilities and reduced the oil loss [169].

The type of lipase (1,3-specific lipase from *Rhizomucor miehei* vs non-specific *Candida antarctica* lipase B) also affected the structuring ability of the process, which could be attributed to either differing stereo specificities or the overall power of the enzyme to produce partial glycerides. *Rhizomucor miehei* enzyme showed lower enzyme activity (longer reaction time) and substantially reduced capacity for producing MAGs, resulting in reductions of attainable SFC and structuring capability. Rather than enzyme characteristics, enzyme quantity can also affect process time. Higher enzyme concentrations can substantially reduce the reaction time, making it much more feasible for industrial usage. Additionally, lipase could be separated upon completion of an industrial-scale GL reaction and reused for additional reactions [62].

The ability to alter the physical properties of the oil simply by changing the reaction conditions highlighted the high potential of this process for producing alternatives for tropical oils and/or interesterified blends of hydrogenated oils, with improved sustainability and health properties [169].

In follow-up research, GL was performed on a variety of liquid oils such as tigernut, peanut, cottonseed, rice bran, olive, soybean, sesame, canola, high oleic algal (HOAO), and high oleic canola (HOCO) oils using optimized condition (*Candida antarctica* lipase B; 65 1C; 48 h; 1:1 glycerol:TAG molar ratio) [172]. The fatty acid composition (SFA and oleic acid content) of the native oil and thus the composition of partial glycerides formed after GL substantially contributed to the crystalline material and physical properties. No crystalline material at 5 °C was observed in HOAO due to having low SFA contents (2.8%) and more than 90% oleic acid. However, canola oil and HOCO with 7% SFAs and much less oleic acid (63.1% and 72.5%, respectively) achieved a SFC of 8.3% and 18.7% at 5 °C, respectively. Overall, structuring ability was highest when native oil with high oleic acid content (>60%) contained more than 10% SFA (ideally 14–25%). In most systems, oleic acid, as monoolein and diolein was responsible for the structuring potential as observed in tigernut, peanut, olive, and HOCO. While monopalmitin was the major MAG species in cottonseed, soybean, and rice bran systems [172].

In a recent unpublished work from the Marangoni group, the GL process of different plant oils was modified and scaled up. Product characteristics were comparable at laboratory and pilot plant scales, supporting the commercial viability of the process. In addition, the potential of structuring oil systems containing more saturated fat such as shea olein and palm olein was investigated for the first time for the purpose of shortening and animal fat mimetic production.

Shea olein and palm olein GPs displayed high plasticity and broad melting peaks with high melting temperatures, similar to animal fat. GPs were then converted to oleogels, using EC with different molecular weights (20 cP, and 45 cP) at 5% w/w to emulate desirable rheological and textural attributes of adipose tissue. A combination of GL and oleogelation produced a more deformable and cohesive product. Comparison of the melting, rheological and mechanical properties of the EC oleogels with those of whole pork, beef, and lamb adipose tissue showed the potential for EC oleogels of oil GPs to be used as adipose tissue mimetics in the new generation plant-based meat analogs.

3.4 Food Applications

Previous works on organogels have been predominantly focused on understanding fundamental properties including microstructure, polymorphism, and rheological behavior [31, 34, 35]. This understanding is critical to applying organogels to food systems as the behavior of traditional fats needs to be mimicked in order to impart similar if not equal functionality in the final food product. In addition, “drop-in ingredient” replacement is highly preferred in the food industry to avoid incurred costs. The desired functional characteristics of the fat content are variable depending on the specifications of the food application. Replacing highly saturated hard stock fats with liquid oil often causes increased oil migration resulting in oil leakage and other quality defects. In response to these problems, an increasing amount of research has focused on exploring the potential use of various types of organogels in a wide range of food applications including spreads, bakery and pastries, cream fillings, ice cream, meat, and dairy products which is overviewed in the following.

Plant wax in soybean oil organogels was used for margarine production, including CLW, RBW, and SFW. It was found that although CLW and RBW were able to form stable organogels, in a margarine formulation CLW showed phase separation and RBW had low firmness; however, SFW was able to form the most desirable margarine characteristics of the three waxes studied [174].

Margarines made using CRW and monoglycerides in virgin olive oil at 3, 7, and 10 wt% concentration levels were studied, all samples, in particular the 7% monoglyceride sample, resembled the textural and thermal properties of a commercial margarine [175].

Shellac organogels were able to fully replace a commercial hydrogenated vegetable oil stabilizer and partially replace (~27%) palm oil in a chocolate paste, and create an emulsifier-free margarine with up to 60 wt% water incorporation [176].

Margarines made using virgin olive oil organogels containing 5 wt% of BW or SFW demonstrated consumer hedonic liking scores of appearance, odor, flavor, and spreadability above neutrality, demonstrating consumer acceptability of these margarine replacements [120].

Structured MAG gels, or Coasun™, are used as an all-purpose shortening replacement in cookies. Traditional all-purpose shortening had superior shortening functionality compared to MAG gels; however, incorporation of the unstructured components of the emulsion resulted in lower cookie dough firmness and increased spread value when compared to its structured emulsion counterpart. Therefore, the structured emulsion maintained a similar dough firmness to the all-purpose shortening dough, indicating that the structured emulsion had a greater ability to act as a structural component in the cookie dough [177].

The ability of MAG gels to act similar to a shortening such as development time and prevention of gluten aggregation was demonstrated [135]. These characteristics are beneficial for baked products that do not rely on gluten network formation for structure, for example, cookies, pie crusts, and pastries.

Traditional shortening was replaced with MAG gel, modified with added 3 to 5 wt % EC [178]. Oil loss was monitored at 37 °C using filter papers to promote oil loss; it was found that cookies made with added EC resulted in lower oil loss overtime than the structured emulsion.

A standard cookie mixture was used to compare cookies containing approximately 24% fat content from organogels prepared with 5 wt% SFW and BW in hazelnut oil and a commercial bakery shortening [48]. Cookies prepared with organogels had a lower aeration (indicated by higher diameter: thickness ratio), lower hardness, higher factorability, and equal or higher positive sensory attribute scores than the cookies prepared with commercial bakery shortening. Therefore, cookies prepared with wax organogels can be considered to exhibit good sensory quality. This was supported by consumer hedonic liking data, which indicated that the organogel cookies were preferred and better accepted than the control cookies [48].

Candelilla wax (3 and 6 wt%)- canola oil organogels resulted in cookies that had more spread upon baking, and lower snapping force contributing to softer eating characteristics than shortening control cookies [179].

MAG organogels were prepared with sunflower oil and were used in sweet bread [180]. Incorporation of MAG organogel resulted in an inhomogeneous lipid distribution in the dough, contributing to low leavening and firmer structure than a palm oil control.

An emulsion-templated structured oil was made with MC and XG as a replacement for traditional shortening in cake batters. It was found that the batter properties and cake attributes of the structured oil were comparable to commercial shortenings and showed significant differences from liquid oil controls [181].

Shellac wax-based emulsions were used as a cake margarine replacement in a basic sponge cake recipe. It was found that though the shellac emulsion batter properties were not comparable to the control in terms of density and flow behavior, the resultant baked cakes showed mostly comparable textural and sensorial properties [176].

A model cream was prepared using a mix of 60% organogel or oil and 40% intersterified hydrogenated palm oil [178]. The organogel was prepared with 6% EC and 2% sorbitan monostearate with either canola or HOSO. An oil binding test conducted at 20 °C using filter papers to promote oil loss showed that creams made with canola oil and HOSO organogel had very low oil loss compared to creams made with oil, demonstrating organogelation is an effective way to slow if not prevent oil migration in cream fillings.

A standard ice cream mix was prepared using 10% RBW in HOSO organogel to replace the solid fat content traditionally used in ice creams such as milk fat, or non-dairy sources such as palm kernel oil, palm oil, coconut oil, or hydrogenated oil. The RBW organogel was successfully emulsified in the ice cream mix and resulted in characteristics similar to that of crystallized fat droplets than of liquid oil. However, the sufficient structure was not developed to cause a delay in a structural collapse during meltdown [45].

Replacement of saturated fat with vegetable oil in comminuted meat products, such as frankfurters, resulted in products that were nearly 3x chewier and had an unacceptable rubbery texture [143, 182]. Frankfurters made with organogels of 10% EC in oil were not significantly different in hardness or chewiness to the beef fat controls [143, 178]. Pork fat in breakfast sausages was also replaced with EC organogels (~20% of the product); it was found the textural qualities of the pork fat control could be matched [183].

Organogels composed of a mixture of RBW, EC, and vegetable oil were used in a cream cheese product giving comparable spreadability, hardness, and had an approximate 25% reduction in total fat content than the full-fat control product [184]. Similarly, a mixture of organogelators in soybean oil was used to make a processed cheese product [185].

Successful application of GL-structured oils in the formulation of margarine and peanut butter has been reported [169, 172]. Emulsifier-free margarine produced with tigernut and cottonseed oils GPs displayed a smooth force-deformation curve like that of commercial margarine, indicative of plastic flow behavior. This was attributed to the thermal softening behavior of these systems, showing high SFC at 5 °C and gradual reduction of solids with an increase of temperature [169, 172]. The authors also suggested rice bran, olive, and HOCO glycerolized systems as other viable options for trans-free spread applications [172]. Similarly, peanut butter made with peanut oil GP presented high stability and comparable firmness to the commercial one [169].

3.5 Formulation Considerations

Whey protein isolate-xanthan gum complexes (WPXC) were used as a fat replacement to partially replace shortening (50 or 75 wt%) in cake frosting and sandwich cookie filling [186]. Response surface methodology was used to optimize the low-fat formulations based on WPXC viscosity and textural properties. WPXC cake frosting and sandwich cookie filling textural, rheological, and melting profiles similar to those of full-fat controls were achieved; however, there were difficulties with water activity for the latter application. The low-fat WPXC had a water activity (A_w) of 0.82 ± 0.02, this A_w is suitable for cake frostings; however, commercial sandwich cookie fillings had an A_w ranging 0.4–0.53 [186]. Since cookie biscuits are hygroscopic, expand with moisture uptake, and have an A_w between 0.2 and 0.6, the moisture would migrate to equilibrium, resulting in the separation of the cookie-cream interface, and a moist cookie without the desirable crisp texture [186, 187]. This is also a problem with cream fillings formulated with highly saturated hard stock fat and unsaturated oils, where migration of liquid oil leads to softening of the cookies and textural defects [178]. In the case of the cream with significant water content, humectants and bulking agents may be used to maintain a low A_w, and for a high-fat-content cream, a suitable structuring agent capable of slowing or preventing oil migration is required. In addition, there are textural differences among cake frostings and sandwich cookie fillings that need to be considered. A highly adhesive product is undesirable because it would have a sticky mouthfeel, a highly viscous product is difficult to pump and extrude during processing, and a fluid-like product would not be suitable to maintain its shape on the product [186]. Soft textures are optimal in the case of cake frostings in order to be spread thinly without breaking the cake crumb, whereas, firmer textures are more desirable for sandwich cookie fillings in order to prevent cookie misalignment, smearing, and shape maintenance, such that cream does not squeeze out when handled or bitten [186, 188]. It is also advantageous for cookie creams to rapidly solidify after spreading to prevent these defects during packaging, storage, and transportation [189].

Starch-lipid composites (SLC) have been a common replacement for the fat content in high-fat foods as the starch component imparts texture and viscosity and the lipid component provides the taste, melting properties, and mouthfeel characteristic of full-fat products [190]. SLC was used as a shortening replacement in cake icing; the SLC samples had hydrogenated fat contents of 1.2–12.8% fat and a constant 0.85 A_w, compared to the 21.5% fat content and 0.84–0.85 A_w of the shortening control. It was found that 16% and 24% SLC content can be used to prepare cake icings with as low as 6% fat content with similar characteristics as the shortening control. Melting behavior of the SLC icings was observed at higher temperatures than the shortening control icing, indicating that these low-fat icings would hold their shape in the mouth when consumed instead of melting like the shortening control [190]. This high-temperature melting behavior, although advantageous for shelf life, may result in a waxy mouthfeel.

3.6 Conclusions

Oleogels provide both environmental and health advantages to animal fats. Most successful food applications thus far involve only partial replacement of animal or highly saturated fats; however, a significant reduction in these fats can still be seen without significantly altering sensory properties. There is a gap in the laboratory structuring of these gels and the commercialization of them. Table 3.2 Summarizes

Table 3.2 Summary of promising direct and indirect methods to structure edible oils

Method	Key points	Limitations	Promising food applications
Wax oleogels	Low concentration required	Waxy taste Time-dependent change of rheological properties Sensitive to water in system Low firmness	Cookies [48, 119] Confectionary fillings [191] Ice cream [45] Cream cheese [184] Spreads and margarine [174, 176] Frying medium [121] Olive oil spread [118]
Fatty acids Fatty alcohols	Similar solid-state structure to TAGs High stability	High amount needed	Margarine [126] Peanut butter [192]
Monoglycerides	No restrictions as food additives Economic Forms crystalline network at low concentrations	Unstable Strength of network is limited compared to TAGs	Emulsifiers in many food products Cake batter [193] Low fat shortening [177] Frankfurter sausage [88]
Diglycerides	No restrictions as food additives Economic	Low crystallization rate Strength of network is limited compared to TAGs	Oleofoams [101–103]
Ethylcellulose	Easy manipulation of gel strength Serum TG lowering effect on oil	Synthetic ingredient Off flavor Limited concentrations permitted	Replacing animal fats in meat products [143, 147, 194] Ice cream [178] Cream cheese [144] Margarine [126] Heat-resistant chocolate [46, 47]
Emulsion template	Can use with biphasic food products Clean label Low gelation concentration	Protein oleogels tend to increase hardens of the final product Limited sensory studies reported	Butter [195]
Foam template			Cake [196] Muffins [197] Sandwich cookie cream [198] Stabilized Peanut Butter [199]

the promising oleogel strategies and their food applications. Further research needs to go into consumer acceptability, economic feasibility, and digestion of oleogels; however, they provide great promise for improving the health and environmental impacts of the food system for the future. On the other hand, enzymatic oil structuring through GL carries the added benefits associated with partial glyceride consumption and commercial viability. So far, GL structured oils are successfully utilized for margarine and peanut butter production. There is also great potential for these products to be used as shortening and as the lipid source in meat analogs.

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