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Advances in Multi-component Supramolecular Oleogels- a Review

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ABSTRACT

Oleogels, with solid-like properties, are made of edible liquid oil entrapped in structures that are produced by oleogelators. In multi-component supramolecular oleogels, due to the positive interaction between gelators, the presence of more than one structurant, causes some advantages in oleogel properties, which make it legally, economically, and industrially usable. A wide range of studies based on multi-component oleogels are focused on the two added components, which are the simplest ones. In this review, three general classes of this kind of oleogels consisting of multi-component gel-phase, multiple gelator components with two sub-groups (co-gels and self-sorting gels) and gelator plus non-gelling additive are examined.

KEYWORDS

Multi-component oleogel; oleogelator; surfactant

Introduction

Lipids play a very important role in our diet because they are the source of energy; lipid-soluble-bioactives such as mono-unsaturated fatty acids, carotenoids, specifically β -carotene; essential fatty acids; fat-soluble vitamins A, D, E and K; as a precursor of steroid hormones, hormone-like compounds (eicosanoids) and vitamins as well as providing satiety.^[1–4] They also have some technological functions such as leaven batters and doughs, improve flavor and texture, contribute to flakiness and tenderness, transfer heat, for example in frying and reduce the length of protein strands mechanically by coating the platelets; moreover, they limit the development of gluten and subsequent toughness by reducing the protein contact with water, stabilize the loaf gas bubbles and prevent sticking in food preparation. They have a desirable impact on the sensory characteristics of food products such as appearance, taste, mouthfeel, lubricity, and flavor, e.g. the richness taste in whole milk, the smoothness of top quality ice cream and desirable sensory sensation of chocolate while being melted at body temperature.^[4–6] According to the recent reports, about 90 million tons of lipids are used for dietary purposes, and 20 million tons are used for deep and shallow frying. Moreover, they are used in spreads including butter, ghee (concentrated butterfat), margarine and Vanaspati (vegetable ghee), baked products (bread, layered doughs, cakes, cookies, short and puffy pastry), salad oils, chocolate, confectionery, and dairy products.^[6]

Based on the physical properties affected by the composition and position of fatty acids on the glycerol molecule, lipids are categorized into fats and oils, which are solid and liquid at room temperature (21.1–23.9 °C or 70–75°F) respectively. Beside health and nutritional benefits of oils (with high liquid to fat crystal ratio), their usage causes some technological problems in the food manufacturing industry such as oil loss, textural defect, and lower shelf life in snacks, chocolates, biscuits, and pastries. Accordingly, hydrogenation, as a successful commercial method, has been used for at least one century in the food industry to improve the technological properties of different types of oil and fat. In this process, hydrogen is added at points of double bonds of unsaturated fatty acids in the presence of nickel catalyst. This process increases the oil stability against oxidation and flavor reversion and improves its properties for specific uses such as deep-frying and baking. However cis-trans isomerization and a high level of saturated fatty acids in the hydrogenated products damage human health. According to the clinical results, the consumption of saturated and trans fatty acids has been linked to obesity, diabetes, high blood cholesterol, cancer, cardiovascular diseases, coronary heart disease, and perhaps gallbladder diseases. Thus, the recommendation of the World Health Organization (WHO) is the intake reduction of trans fatty acids and supplementation of 10% of the required energy from fats.^[1–3] Regarding this regulation, researchers make an effort to structure kinds of liquid oil using novel methods. They attempt to eliminate or reduce trans and saturated fatty acids in food products, to omit palm oil from the formulation of products due to consumers' environmental concerns and to eliminate technical problems such as textural defects and oil leakage.^[2,7]

In this regard, the organogelation concept, gelation of oil, has recently been offered in the pharmaceutical, food, biotechnological, cosmetic, and petrochemical industries. The edible-oil gels are called oleogels to differentiate them from other organogels. oleogel, which displays solid or solid-like behavior, can be used as an alternative to traditional fat to demonstrate desired functional properties. In contrast to the conventional technique, the oleogelation process is a simple and low-cost method which does not have any effects on the oil fatty acid structure. Actually, 3-dimensional thermo-reversible network, which is constructed by oleogelator, imprisons liquid oil.^[3,8–11] Oleogelator should be natural, active on surface, food-grade, self-assembly, thermo-reversible, easily available, economically viable and cheap. Moreover, it should give a higher quality at lower concentration, preferably at 0.5–2% (weight bases).^[3,12–14] The rheological, textural and thermal properties of oleogels can be affected by oil characteristics (type^[3,8,15–17], polarity^[3,16,18,19], oxidation state^[8] and viscosity^[16]), structuring agent features (type^[18], composition^[18,20–23], concentration^[15–17,24–26] and surfactant addition^[17,27]), process conditions (cooling rate^[8,18,26,28] and shear rate^[18,29]), storage situation (time^[18,20] and temperature^[20–22]) and finally the crystal network microstructure.^[21]

The classifications of organogelators are based on the features of organogelator molecules, their chemical characteristics, and the kind of their building blocks. Based on the features of organogelator molecules, they can be classified into low-molecular-weight organogelators with less than 3000 Da, polymeric organogelators, and inorganic compounds. N-alkanes or paraffins, free fatty acids and fatty alcohols and mixtures of them, waxes, triacylglycerol (TAG) derivatives, phytosterols (PSs), ceramides, surfactants, lecithin, shellac waxes, amides and finally sugar-based gelators are low-molecular-weight

organogelators. Cellulose derivatives (ethylcellulose (EC), hydroxyl-propyl-methyl cellulose), proteins, and water-soluble polysaccharides are high-molecular-weight gelators. Fumed silica is an example of an inorganic compound.^[2,3,10,30] According to the chemical characteristics, there are lipid and non-lipid based gelators. Most of the gelators, which have been mentioned above as low-molecular-weight, are placed in lipid-based ones, whereas polymeric organogelators, EC, and proteins are non-lipid-based gelators.^[31] Based on the kind of building blocks and basic principles of structure, organogelators are divided into four groups as follows: Crystalline particles, which can be mono-component (natural waxes, mono, and di-acylglycerols) or mixed systems (fatty acids+ fatty alcohols, PSs+ monoglycerides (MGs), phospholipids+ sorbitan esters), comprise the first group, while self-assembled structures of low molecular weight compounds, which can also be mono-component (self-assembled fibrous network forming fatty acids, ricinoleic acid, 12-hydroxy stearic acid, and sphingolipids) or mixed systems (PSs+ sterol esters, phospholipids+ tocopherols), are included in the second group; the third group is self-assembled structures of polymers or polymeric strands, which can be produced by either direct (EC) or indirect (methylcellulose, hydroxyl-propyl methylcellulose, gelatin and protein+ polysaccharides) methods, and finally the last group is miscellaneous structures such as inorganic particles (fumed silica) and emulsion droplets (high internal phase emulsion gels).^[32]

A variety of gelators, which can structure vegetable oil individually, have been identified, and their oleogel properties have been widely investigated. However, one of the major problems is their similarity with specific functional properties of fat. Gelatos' combination, which has not yet been investigated completely, can be effective in broadening the gel properties. Accordingly, oleogels can be mono-component or multi-component. A single gelator is used in mono-component oleogels, whereas Multi-component oleogels consist of more than one gelator. The simplest multi-component oleogels, which are widely studied to date, have two gelators. Compared to the mono-component, it is easier to tailor the features of the gelator in the multi-component type of oleogels to the application and functional properties.^[27,33–36] Additionally, all the essential and technological qualities and consumers' desired properties in hybrid systems can be achieved more successfully. Moreover, overcoming the regulation issues about the gelator concentration used in the food industry can be achieved by using the synergistic effects of gelators.^[12,18,30,37–40]

Three different groups of multi-component oleogels (Fig. 1) are as follows:

- (1) Two or more components are required for gelation, and each component can't form gels individually.
- (2) More than one gelator is used to form oleogels, and each gelator is capable of forming structured materials. This category is divided into two subclasses:
 - (a) Co-assembled gels include gelators which interact with each other to form a gel.
 - (b) Self-sorting gels, where assembled structures are formed by each gelator independently.
- (3) Gelator(s) and non-gelling additive(s) are added to the edible oil to improve the gel properties.^[27,33,34,41]

In recent years, many beneficial books and review articles have been focused on the classification and properties of the structuring agent, oleogel production methods, their



1- Two component gel-phase

Where each component can't form gels individually



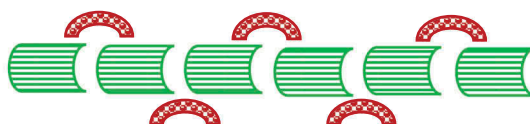
Or



2- Two gelator components

2-1- Co- assembled gels (co- gels)

2-2- Self- sorting gels



3- Gelator plus non-gelling additive component

Figure 1. Schematic representation of the different classes of multi-component supramolecular oleogels. Adapted from Patel.^[27]

characterization and food application, factors affecting the gel behavior, and specific gelator-based oleogel features.^[3,8,9,11,18,27,34,38,39,41-47] However, none of them has paid attention to the multi-component supramolecular oleogels perfectly. Regarding the benefits of this issue in the food industry and its necessity for a comprehensive review, the objective of this study is to look at the recent researches of multi-component supramolecular oleogels thoroughly.

Multiple component gel-phase

Lecithin, sitosterols, sorbitan tristearate (STS), α -tocopherols, and sucrose oligo-esters with Hydrophilic-Lipophilic Balance (HLB) 2 are examples of components that can't form gels individually.

Lecithin plus other component(s) gel-phase

Lecithin (phosphatidylcholine), as a component of non-polar materials, is a natural surfactant used in the food, pharmaceutical and cosmetic industry. It has a small hydrophilic head and two large hydrophobic tails (Fig. 2a). According to the fatty acid chain bonds, lecithin can be found in the saturated or unsaturated forms. Compared to saturated lecithin, the only unsaturated type can act as a plasticizer in oleogels. Lecithin creates small spherical reverse micelles in a non-polar solvent [Fig. 3a]. Due to the formation of strong hydrogen bonding, the addition of the small amount of water or a polar solvent changes the molecular packing parameter. This converts the geometry of micelles to reverse cylindrical micelles or lamellar phases [Fig. 3b] and creates supramolecular polymers or supramolecular structures. Subsequently, a three-dimensional network develops to trap liquid oil via overlapping. If the polar solvent is water, strong interactions protect network up to 65°C.^[48,49] Lecithin affects the self-assembly of other surfactants and is used in combination with α -tocopherol, STS, and sucrose esters (SEs).

Purified lecithin, which combines in 1:1 ratio (w/w) with α -tocopherol in sunflower oil, can produce oleogel at 25% total gelators concentration, while these components can't form gels in any vegetable oil at any concentration individually. The size differences in the hydrophilic and hydrophobic area of lecithin and α -tocopherol (Fig. 2) decrease the molecular packing diameter and construct cylindrical micelles which create an oil trapping network via overlapping. This network is shear and temperature-sensitive, while the addition of water not only doesn't weaken but also strengthens it up to 1.5 times. The lecithin/ α -tocopherol organogels (with or without water) can be used as crystalline fat both in edible stuff, and pharmaceutical, and chemical industries.^[48]

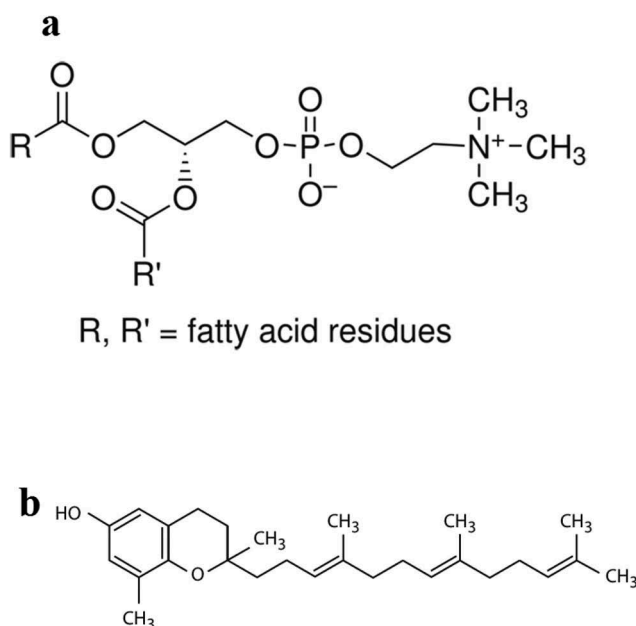


Figure 2. Molecular structure of (a) phosphatidylcholine and (b) α -tocopherol.

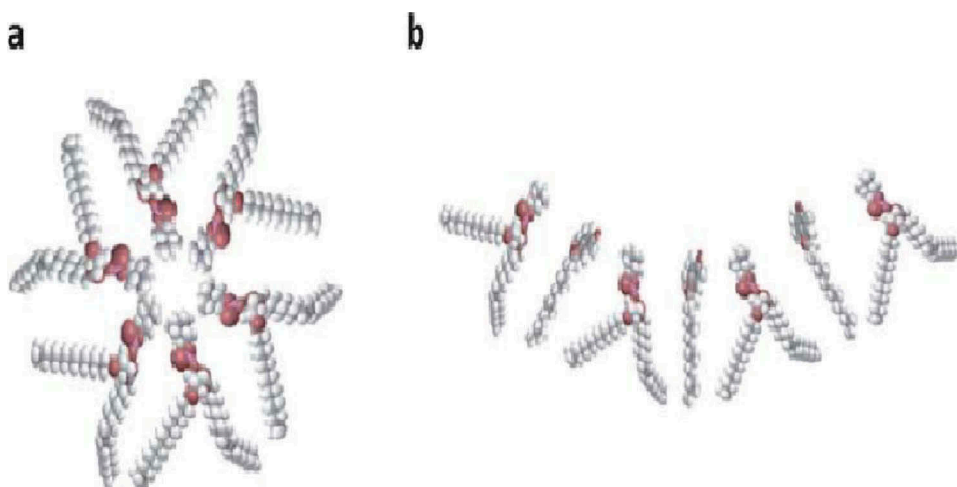


Figure 3. Schematic representation of the packing geometry. (a) Inverse micelle formation with large curvature (b) formation of inverse cylindrical micelles or lamellar phases. Adapted from Nikiforidis, and Scholten^[48].

The structure and crystalline form of lecithin/STS oleogel are given by STS at specific ratios between 40:60 and 60:40. In these oleogels, lecithin prompts the formation of needle-like or plate-shaped crystals and the connection of crystals by weak junctions to form a network that entraps liquid oil. The oil binding capacity (OBC) of oleogels improves during storage time. However, in comparison with glycerol monostearate (as mono-acylglycerol (MAG)), the critical gelation concentration of lecithin/STS is higher, and its melting point, OBC, and hardness are lower than those of oleogel of MAGs in refined soybean oil (SBO). In gist, as a result of thixotropic qualities and zero solid-fat content at 40°C, lecithin/STS oleogels can be used in confectionery products to prevent oil migration without producing waxy taste. These oleogels have a good ability to flow during pumping in the food production process.^[23,50]

Sugar esters are amphiphilic surfactants with different fatty acid compositions and HLB values. In SEs, which are non-ionic surfactants, fatty acids are esterified with sucrose hydroxyl groups. Based on the number of esterified groups, there are mono, di, tri, and up to poly-esters of sucrose with different values of HLB, which determine their usage for oil in water (O/W) or water in oil (W/O) emulsions. In oleogels containing SEs (HLB 2) and sunflower lecithin (SFL) in sunflower oil, SEs participate in the production of building blocks, and SFL signifies the SEs aggregation by getting involved in hydrogen bond formation between SEs monomers, while the mono-component of these structurants can't form gels. The resulting oleogel has better structural formation and mechanical properties at 7:3 ratio (SEs: SFL). This information is valuable for greater application of SEs in food and pharmaceutical products.^[51]

Other(s) multiple component gel-phase

The mixtures of unsaturated fatty acids and sodium fatty acid salts can produce a network in aqueous environments. In this matter, sodium fatty acid salts are the only cationic fatty

acid salts, which assist in the network formation via alteration of spherical micelles to cylindrical micelles. The length of fatty acid chains is also the other important factor. Accordingly, though oleic acid (HLB~1) and sodium oleate (HLB = 18) can't structure liquid oil individually due to their synergistic interaction, their mixture (at above 10% w/w and in 1:1 ratio) in refined sunflower oil gives an oleogel with a very fine structure. In this oleogel, oleic acid inhibits the considerable growth of sodium oleate aggregates, and the presence of sodium oleate causes the structural alteration of oleic acid from reverse spherical micelles to lamellar crystalline structures. Since water facilitates the hydrogen bond formation between sodium oleate and oleic acid, the gel strength increases due to water concentrations below 1.5%wt. However, the hydrophilic nature of sodium oleate leads to phase separation at higher concentrations of water.^[52]

Multiple gelator components

In this category, two or more components, which can structure liquid oil individually, are used together. These gelators either assemble together (co-assembly) or remain independent of each other (self-sorting). Co-assembled gels are the widest group of multi-component oleogels in research studies.

Co-assembled gels or Co-gels

In general, co-gel is more effective than pure components.^[39] Some examples of two structurant mixtures are fatty acids+ fatty alcohols, sterol+ sterol ester, and fatty acid + sorbitan ester co-assembly.^[27]

Sorbitan esters based Co-gels

The crystallization behavior of fats can be changed by sorbitan esters of fatty acids.^[53] Accordingly, the generation of a more stable form of fat crystals is delayed because of the presence of sorbitan monostearate (SMS or Span 60) and STS. In this context, the hydroxyl groups of the polar head of span 60 interact with the free hydroxyl groups of monodiglycerides or beeswax ester groups. This inhibits the joining of saturated side chains of monodiglycerides or beeswax (BW), and it also prevents the formation of tubular structures; subsequently, it decreases the crystallization temperature and makes the gels more elastic. Pérez-Monterroza et. al.^[24,25] asserted that this negative effect of SMS in combination with either monodiglycerides or BW could be positively used to control the crystallization temperature of avocado oil oleogels. The crystallization of SMS caused the heterogeneous nucleation of BW. Oil fatty acid composition, structuring agents' concentration, and kinetic effect of cooling rate influenced the formation of different β or β' crystals in oleogels.

Stearic acid (SA) and its derivative (12-hydroxy stearic acid) are used as structuring agents for various kinds of vegetable oil. Apart from the crystallization rate, sorbitan esters of fatty acids (the trade name of Span) and polyoxyethylene sorbitan esters (the trade name of Tween) can also affect the presence of the SA specific polymorph. Correspondingly, an increase in the firmness, a decrease in the nucleation induction time, and incitement of the secondary crystallization of SA can be obtained in the presence

of SMS. Moreover microstructural properties of SA oleogel change from plate-like structure to branched structure in the presence of SMS. Additionally, the formation of polymorph A (mesh-like structure) and heterogeneous nucleation of SA crystals occur due to the presence of Span 80 in SA oleogel. SA/Span 80 containing oleogels also show the secondary crystallization, heterogeneous nucleation, and higher firmness. Based on the results, the SA/Span 60 multi-component co-gel will have higher diffusion and fluidity than the pure SA oleogels. These properties suit them to spreadable product formulations and drug delivery services.^[53,54]

Sterol-sterol ester co-gels

PSs, which are derived from plants with a similar structure to cholesterol, can reduce cholesterol absorption and, subsequently, decrease the level of low-density lipoprotein (LDL)-cholesterol (bad cholesterol). In TAG systems, phytosterol (PS) crystallization is prevented or reduced owing to the existence of lecithin as a solubility aid and MAG as a trap which is made of a dense crystalline network.^[55] β -sitosterol and γ -oryzanol oleogels are one of the most common, approximately food-grade, and important multi-component systems in scientific researches. In this system, the assembled double-walled hollow tubules, with temperature formation higher than the other ratios, build a network in a 1:1 molar ratios (2:3 wt% β -sitosterol: γ -oryzanol). From the thermodynamic point of view, this process is thermo-reversible, because the negative value of Gibbs free energy of tubules formation. The crystal nucleation, which is purely instantaneous at 10% w/w (the Avrami growth parameter, $n = 0.9$), changes into sporadic at 20% w/w ($n = 1.6$) and subsequently modifies the structures. The main structure is built by sterol molecules, while acyl-chain and a ferulic acid moiety of γ -oryzanol stick out from the fibers' wall and prevent them from becoming close to each other.^[16,56-59]

The physical properties and self-assembly kinetics of PS/PS ester oleogels are influenced by the sterol mass ratio. At 60:40 (oryzanol: PS) ratio, the oleogel is elastic and transparent. At higher PS content, the crystallization of excessive amounts of PSs increases the opacity and brittleness of gel. The physical properties are also directly time-dependent.^[60] Inter-fibril non-covalent linkages among methoxyl, hydroxyl and aromatic groups of ferulic acid are necessary to produce a strong network.^[61] The oscillating shear facilitates the gel formation rather than the quiescent condition.^[62] Oil viscosity is the main factor in the gelation process of β -sitosterol and γ -oryzanol. Whereas the nanostructure of oleogel has not been affected by the oil type, as a result of difficulty pack together, fewer structurants exist in the mixture as the oil viscosity increases.^[16] Additionally, oil polarity influences the appearance and thermodynamic behavior of organogel. Accordingly, higher polarity increases the sterol solubility, gel transparency, thermodynamic parameters (enthalpy, entropy, and Gibbs free energy) and instability of gel against temperature changes.^[63] Whereas the size, shape, and number of crystals, oil retention as well as the textural properties (firmness and adhesiveness) are directly related to the gelator concentration, the thickness of tubules is not affected by it.^[60,64] The structure of tubules is more affected by the presence of water. The substitution of water with a polar phase such as glycerol can produce oleogel-glycerol emulsion, which can be used in low-fat confectionary products.^[65,66]

Other sterols such as ergosterol, stigmasterol, cholesterol, and cholestanol (Fig. 4) can substitute for sitosterol in the tubules.^[67] Dalkas and his coworkers^[61] demonstrated that hydrogen bonds between hydroxyl and carbonyl groups of PS and sterol-esters respectively; in addition, the assemblage of the stearine group of PS with the stearine group of sterol-esters, is necessary to form stable tubules. Therefore, the formation of tubules is based on the presence of the acid moiety in the sterol ester.^[67] The presence of a hydroxyl group and double bonds in the ring system of these sterols affect the gel formation, while the chemical structure of the alkyl chain has a little effect on the ability of gel formation.^[62] The findings show that sterol/sterol ester oleogels can be applied in the filled chocolate product as oil migration barrier^[7], commercial shortening^[26], cooked food products such as high-fat sausages and frankfurter sausage^[64,68], and lipophilic delivery systems.^[64] Accordingly, the substitution of organogels prepared by mixing γ -oryzanol and PS in sunflower oil for 10% pork backfat in frankfurter sausage, with a few minor exceptions in color, doesn't change the pH, degree of lipid oxidation, textural features such as hardness, gumminess, and chewiness and sensory properties including appearance, smell, taste, texture, oiliness, juiciness, and overall acceptability of the final product significantly.^[68]

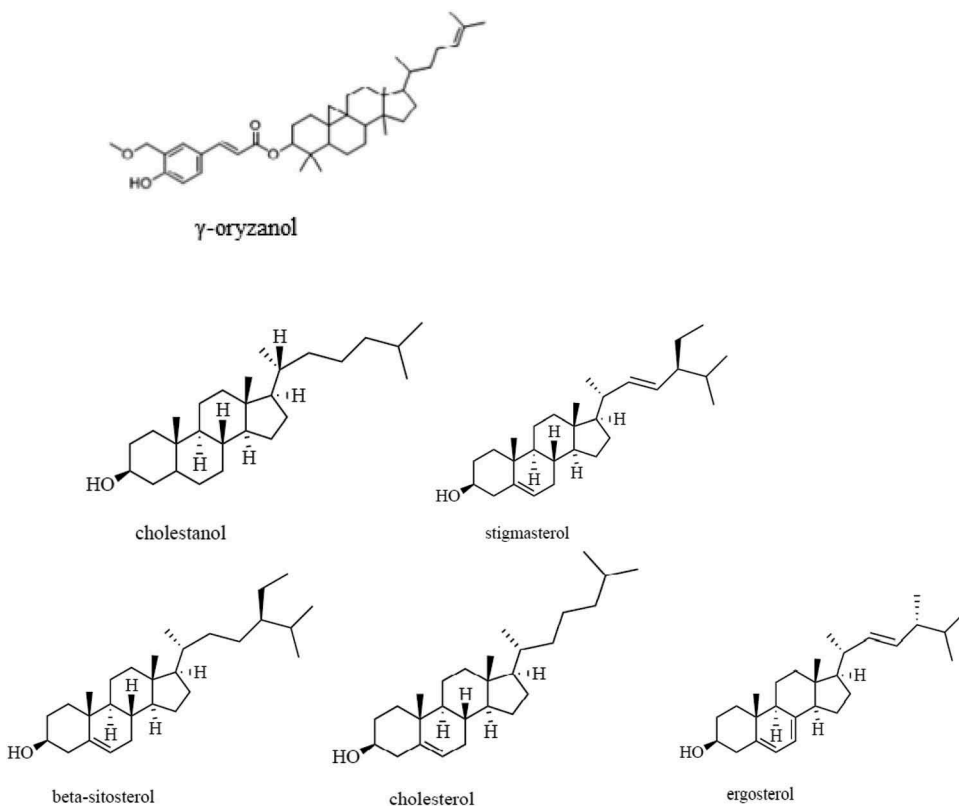


Figure 4. Chemical structures of γ -oryzanol (top) and sterols.

Phytosterol-mono-glyceride co-gels

Due to agglomeration and sedimentation, phytosterol based mono-component oleogels are not stable.^[69] Moreover, native phytosterols (NPSs), as plant-derived sterols and stanols, cause sandy and gritty texture in food products due to the high melting point. They also can't structure oil individually, even at 10% by weight.^[28] On the other hand, MGs mono-component oleogels, which suffer polymorphic transition on aging, require high concentration gelator, and are sensitive to processing conditions.^[70] Consequently, researchers have tried to modify their assemblage using molecular and crystal engineering.^[69] Accordingly, the combination of either MGs or PSs and other gelators, for example, PS esters^[16,56,60-65], lecithin^[20-22,71,72], waxes^[73] and EC^[74,75] as well as, MGs-PSs co-gels^[28,69,70,76,77] was investigated to improve the gel properties. In this context, glyceryl monostearate (MGs) presence in PS-based oleogels forms a stable gel, with renewed small spherical crystals in the double-layer ordered structure.^[69] Sintang et. al.^[70,76] related the improvement in physical stability of MGs-commercial PSs oleogels, at an 8 to 2 ratio and 10% of total concentration, to the formation of needle-like crystals and reduction of clustered crystals content. In consequence of PS action, compacted alkyl tails of MGs improved viscoelastic properties. Compared to the mono-component oleogels, not only was PSs crystallization hindered, but crystalline clusters and flocs of MGs were disintegrated in the mixed systems, which improved the rheological properties. Moreover, the connection of MGs into the arranged structure was declined by PS. The resulting oleogels with smaller uniform crystals, compared to mono-component gels, had a slower polymorphic transition, longer crystallization time, lower solid-fat content, higher resistance to shear, better structural recovery, and higher strength.^[70,76] The synergistic effect between MGs and PSs at 15:5 (w/w) ratio was more pronounced than 10:10 (w/w) ratio. At a greater amount of PSs (10% of total gelator concentration versus 5%), as a result of the variety in crystal size, the formation of large crystals from excess PSs beside small crystals resulted from the interaction of both gelators, therefore, the network was weakened, while stronger oleogel was formed at 15:5 (w/w) ratio.^[77] With the aim of using the nutritional properties of NPS, researchers try to construct oil foams from oleogels by a whipping action using NPS and MAG. MAG is the main oleogelator in this system and NPS was co-crystallized with MAG. The stability of air bubbles in the resultant oleofoam, whipped oleogel, is created by layer(s) of lipidic crystals around the air bubbles. Since the functionality and application of oleogels have been influenced by MAG: NPS ratios and thermal history, faster cooling rate will reduce the crystal size, complex modulus (less firmness and rigidity), and whipping time significantly.^[28] The application of this co-gel has been investigated in frankfurter sausage formulation at 50% (by weight) replacement for pork back-fat^[77], spreadable low saturated fat foods^[28], butter and margarine.^[70,76] They can also be used as functional ingredient carriers^[70,76] and flavor delivery, especially the lipophilic compounds.^[69]

Fatty acid-fatty alcohol co-gels

Fatty acids and fatty alcohols have the oil structuring ability at 2% (w/w) concentration. However, their combination, namely Palmitic, Stearic, Arachidic, and Behenic acids with Palmityl, Stearyl, Arachyl, and Behenyl alcohols respectively, can structure sunflower oil at a total concentration of 5% and in different ratios. The synergistic effect on the gel hardness is significant at 7:3 and 3:7 (by weight) ratios at temperatures $\leq 20^{\circ}\text{C}$.^[78] In this group, one of the first identified combinations with the highest ability to produce oleogel is stearyl alcohol (SO)-SA mixture.^[79] The beginning structurant concentrations of crystallization and network formation

of SA-SO oleogel are around 1.5% and 3.5% respectively.^[40] The concentration of SA directly affects SO fibers' growth. The polymorphic transition of SO, temperature and speed of crystallization, uniformity, firmness, mechanical stability of oleogel, and network capability for molecular rearrangement are directly affected by SA concentration.^[80] According to the x-ray diffraction patterns, the molecular packing arrangement of SO is influenced by the addition of the small amount of SA.^[79] Modification of the SO network, in the presence of SA, affects the firmness of the oleogel. The higher quantity of SA, the higher firmness.^[80] According to Blach et. al.^[79], the oleogels microstructure is influenced by the oleogel preparation technique. Unlike Schaink et. al.^[40], they have illustrated that the plate-like structures are the only structure in cryo-SEM micrographs of all samples regardless of the composition of structurant. In their opinion, the crystalline mass distribution is the main factor that affects the texture of the gel. Compared to the mono-component, the synergistic interaction of SO/SA in the gel hardness and rheological properties can be observed in 8:2 (in canola oil), and 7:3 (in canola oil, sunflower oil, and high linoleic acid oil) ratios. Accordingly, the oleogel hardness in 7:3 ratio is 2.5 folds higher than the pure components or the other ratios. Stable gels, owing to the high mechanical strength, have the highest rate of OBC.^[40,79] The presence of EC increases the initiation temperature of crystallization and improves the nucleation process of this system in canola oil. The greater efficiency of SA and SO in gelation achieved in the presence of EC. Despite the strong effect on the organization, EC doesn't modify the basic polymorph (platelet-like morphology) of SA/SO crystals. The high ratio of SA or SO (10:0, 1:9 and 0:10) in the presence or absence of EC causes the highest stability, and OBC of oleogels, while at the intermediate ratios (8:2–4:6), the presence of EC weakens gels and increases the oil loss.^[9,36] Alteration of the SA/SO content allows the adjustment of the melting temperature of oleogel in topical applications.^[80] Margarine, halvarine, sausages, and Dutch croquettes have been suggested to contain this type of structurant as fat mimetic in their formulations.^[40] The ability of these oleogels for drug delivery and maintenance of its active form is better than SA oleogels.^[80] SA-SO-EC based oleogel in a definite ratio can also be used in spreadable fat such as margarine.^[9]

Waxes based co-gels

The combination of two different waxes (BW, rice bran wax (RBW), sunflower wax (SFW)) and their ratios in SBO, affect the melting behavior of the oleogels, due to the presence of different components. Wax solubility is probably a determinative factor in crystal morphology. Not only increasing the enthalpy of systems but also the influence of the minor components and mainly the crystallization behavior are affected by the presence of wax esters in the chemical composition of waxes. Solid-fat content values which give some information about real formulated products and their mouthfeel at 25 and 35 °C respectively have shown that SFW and RBW, with higher melting temperature than BW, can cause waxy mouthfeel in binary blends, whereas BW, as a result of varied chemical composition, has a softening effect and causes the formation of eutectic composition. The synergistic effect of RBW and SFW causes the highest storage modulus (highest solid-like behavior) of their oleogel.^[81]

Candelilla wax (CLX), which has been approved as a gelator by Food and Drug Administration (FDA), can gel liquid oils at very low concentrations without any limitations. Its gelation ability is due to the plate-like structure formation by hentriacontane as its major n-alkane. However, the rheological data has confirmed that CLX can't stand high shear stress, and its oleogels are highly shear sensitive. Accordingly, researchers have tried to improve its

physical properties in binary gelator systems. As a result of scientific researches, co-crystallization of CLX with tripalmitin in safflower oil, under the shear rate of 600 s^{-1} and during the cooling stage to nucleation or metastable temperatures, produced an organogel in an orthorhombic subcell with higher elasticity.^[12,29] The other study has shown that, though the presence of MAGs and hard fat together with CLX in oleogels reduces hardness, as a result of co-crystallization of structurants, but it improves the thixotropy properties and the rheological behavior. This reduces the shear sensitivity of oleogel and subsequently, improves its OBC. CLX, MAGs, and hard fat ratios are more important on crystal morphology, texture profile, and all in all physical properties than total gelator quantity.^[12] On the contrary, Toro-Vazquez and his coworkers^[82] believe that the crystallization of MGs and CLX during the MG-CLX-safflower oil organogelation process occurs separately from each other without any molecular interactions. Nevertheless, inter-crystal interactions of oleogel are influenced by MG, and they are especially more significant at higher concentrations of MG.

Other Monoglycerid co-gels

In addition to PS/MG and CLX/MG co-gels, which have been mentioned previously, the other components of MG co-gels are fatty alcohol/MG, EC/MG, and MG/MG co-gels. Besides MGs ability to entrap liquid oils, the high usage of them is influenced by the fact that they are common emulsifiers in foodstuffs and cosmetics, economical and versatile gelators. In glycerol monopalmitate (GMP)/glycerol monostearate (GMS)/virgin olive oil organogel, the inhibitory effect of the ethyl group in GMS against the hydrogen-bond formation weakens the rheological properties, thus the stability of organogel is more dependent on the GMP concentration. However, oleogels with 90% GMS of total gelator concentration are suitable for partition drug release between gastric and intestinal tracts acceptably.^[83] On the contrary, the use of various types of MGs as surfactants in the model chocolate system at a 1:3 ratio with EC, the plasticizing effect of surfactant causes more extension of polymer in the oil and higher hydrogen bond formation between EC and sucrose molecules. This increases the mechanical strength of the system.^[84]

Policosanol (P), as fatty alcohol of RBW, beside the nutritional value, can structure different kinds of liquid oil. In oleogels containing a mixture of glyceryl stearate (GS) and P in virgin olive oil, P determines the crystallization and gelation behavior of the oleogel. In this context, P leads to network formation. Thus, the properties of gel tend to the P oleogel properties, while the strength and consistency of oleogel are influenced by GS crystals. Based on the Fourier Transform Infrared Spectroscopy (FTIR) analysis, resulting in P presence, van der Waals force influences gel formation at low concentrations, whereas hydrogen bonds and van der Waals forces act at a high amount of gelators.^[85]

Self-sorting gels

In self-sorting gels, the pure assembled structures of different gelators are segregated from each other by thermally induced gelation process. On the cooling stage, the assemblage of one gelator molecules at a higher temperature can cause a self-sorting network formation. Detection of self-sorting structures can be inferred by scanning electron microscopy (SEM), transmission electron microscopy (TEM), or atomic force microscopy.^[86]

β -sitosterol-stearic acid self-sorting gels

A combination of β -sitosterol and SA in sunflower oil produces a self-assembled structure, while each gelator maintains its crystalline shape in the network and doesn't interact with each other. Increasing the concentration of gelators, at a constant ratio, results in variation in size and appearance of crystals, which attribute to the differences in solubility and increase in the saturation of the solvent. Variation of gelators in terms of ratio at a constant concentration or vice versa causes the polymorphic transition and rearrangement of molecules. In this system, SA concentration determines the thermodynamic properties of the oleogels. The hardest oleogel can be achieved at β -sitosterol: SA ratios of 1: 4, and this is followed by 0: 1 ratio. The higher firmness of two-component oleogels compared to the pure one is due to the filling effect of β -sitosterol fiber-like network in spaces among the SA crystals.^[87] β -sitosterol and SA also produce self-assembled structures, with thixotropic shear thinning behavior, in extra virgin olive oil, corn oil, and flaxseed oil. According to the results, flaxseed oil with lower viscosity and higher functional properties is the best oil to prepare β -sitosterol-SA oleogel.^[15]

Wax self-sorting gels

To achieve unique oleogels, which are suitable for application in different foods, Tavernier and coworkers^[88] produced oleogels by combining low-melting (berry wax or BEW) and high-melting (SFW and RBW) wax in rice bran oil. They also prepared an oleogel of SFW/RBW in rice bran oil. Based on the results, RBW did not affect the melting and crystallization behavior of BEW, but its crystallization reduced by 1% SFW. The synergistic interaction between SFW and BEW increased the critical stress, cross-over point, and the average of elastic modulus, in the linear viscoelastic region of SFW/BEW oleogel. In RBW and BEW combination, the synergistic interaction, which directly determined the gel hardness, was dependent on the storage time and the oleogelators ratio, whereas, in SFW/BEW combination, the storage time was not effective. Since there wasn't any correlation between solid-fat content and hardness in binary systems, researchers suggested the application of the sintering process, the formation of solid bridges within the pre-existing crystal network, in oleogel hardness. As a result of the crystallization and melting behavior sequentially, β' and γ polymorphs were present together within the mixed systems. The ability of gelators in oil retention and gelation depended on the produced crystal structure and surface area. The lower surface area and crystal interconnection were, the lower the oil binding and gelation capacity stood.

Gelator plus non-gelling additive component

In this group, surfactants such as lecithin and Tween 20 are often used as non-gelling additives.^[89] Spherical, and cylindrical micelles, vesicles, and lamellar structures are shapes of surfactants which are modeled by calculation of critical packing parameter (CPP). Surfactant, as an amphiphilic compound, can structure aqueous and organic medium through self-assembly.^[51] Their functions, as the improver of the stability and plasticizer, modify the mechanical properties of the system.^[89]

Gelator(s) plus lecithin component gels

Soy lecithin at 20 wt% and 30 wt% oleogelator concentration with the incorporation of 1 wt% water can produce oleogel. However, this oleogel is not stable enough to endure shear and high water content. Nonetheless, the presence of SA can stabilize the lecithin oleogel and oleogel emulsion. The stability of oleogels is due to the network formation by reverse micelle fibers of soy lecithin, and their interaction with polar and non-polar tails of crystals and spherulites of SA. SA concentration is a major determinant of thermal behavior, firmness, and accelerated oil loss in oleogels containing both components.^[90]

Due to the reduction of β - sitosterol supersaturation degree, lecithin improves the self-assembling capacity, and thus the oleogel formation ability of β - sitosterol.^[22,71] Accordingly, the presence of lecithin in the β - sitosterol/lecithin/high linoleic acid sunflower oil (*HLSO*) oleogel alters the supramolecular morphology from rosette-shaped (in pure β - sitosterol/*HLSO*) to needle-like structure (with better structuring ability) and increases the crystal-crystal contacts especially at higher storage temperature (T_s). Thus, the hardness value of mixed component oleogel is higher than that of pure sitosterol or lecithin organogels at 25°C. While hardness and storage modulus (G') are directly associated with the sitosterol content at lower T_s (5°C), the synergistic effect between sitosterol and lecithin affects oleogel hardness and G' at higher T_s (15 and 25°C). This indicates that the interaction between gelators increases at higher T_s . Consequently, this mixed structurant system has a higher destructive temperature. According to these results, the authors recommended the sitosterol to lecithin mass ratio of 8: 2 as the best structuring ratio.^[21,22] As a result of the pseudoplastic behavior of β - sitosterol/lecithin/sunflower oil oleogels, they can be used in food formulations to form a thin layer on the surface without any changes in physical properties for at least 60 days.^[20] The resultant oleogel of these two gelators in corn oil, at 8:2 (sitosterol to lecithin) ratio and 12% concentration, can also be used as the curcumin carrier. According to the results, incorporation of curcumin doesn't modify the crystal morphology and hardness of oleogel significantly. The bioavailability of curcumin improves before meals in oleogel structure up to 67.66%. Additionally, the effect of molecular mobility restriction of gel structure increases the oxidative stability of oleogel containing curcumin during the storage time.^[71]

Lecithin functionality as solubility aid increases the solubility of oryzanol and encourages the formation of inverse mixed micelles via intramolecular hydrogen bonds in sunflower oil. As a result of the equimolar mixture of lecithin and oryzanol in sunflower oil, the viscous solution is converted to gel by the addition of about 17% (w/w) water. Due to the similarity in rheological properties, the resultant gel can be used as spreadable fat, with cholesterol-lowering properties.^[91]

Researchers have considered lecithin contribution to oleogel formation either directly or indirectly as an accessory additive. However, in some hybrid oleogels, it has exhibited a disruption effect.^[92] For instance, in a ternary structurant system of β -sitosterol/ γ -oryzanol/lecithin oleogels, lecithin influences the shape and properties of the fibrous structure and makes the gelation process difficult. It also reduces mechanical properties (>20% replacement) and thermal stability, and delays structure shear recovery.^[72] Likewise, incorporation of the polar head groups of lecithin and carboxylic groups of 12-hydroxy stearic acid, in complexes with a 1:1 molar ratio, changes the microstructure of oleogel and inhibit the gel formation.^[92] A similar effect was observed by Aguilar-Zárate

and coworkers^[49], where reducing the interaction and aggregation of EC oleogels beyond the critical concentration of unsaturated soy lecithin reduced the shear moduli. Nevertheless, oleogels structured by unsaturated soy lecithin and EC in high oleic canola oil displayed a comparable thickening response to lard fat.^[49] Lecithin also causes some disruption effects on the production of EC-heat resistance chocolate. Actually, in chocolate formulation, lecithin, other phospholipids, and probably other emulsifiers improve the flow properties by covering the sucrose molecules, so they prevent from the hydrogen bond formation between sucrose-sucrose crystals. However, this effect also hinders the interaction between sucrose crystals and EC, which affects the heat resistance. Thus the mechanical strength and heat stability of the sample are affected negatively.^[84,93] Additionally, lecithin has an antagonistic effect on high-melting-point (SFW and carnauba wax (CBW)) and mid-melting-point (BW and CLX) waxes. Interestingly, the synergistic interaction of lecithin with low-melting-point waxes (BEW and fruit wax (FW)) reduces the critical gelling concentration of wax to produce an oleogel in sunflower oil. This synergistic effect is related to the similarity of carbon chain length between FW, lecithin, and sunflower oil. Crystallization is due to the wax action, and lecithin strengthens the gel structure by self-assembly and organization of the crystal network (up to 50% w/w of FW replacement).^[37]

Wax plus surfactant component gels

Wax oleogel application as a solid-fat replacer in ice cream formulation has been investigated in some researches. Moreover, glycerol monooleate (GMO), as a usual surfactant in ice cream formulation, creates a membrane at the oil/water interface. In addition to the wax type and the emulsifier kind, the oleogel structure in ice cream is improved by the oleogel concentration and high-shear freezing process. Compared to the other waxes (RBW, CLX, CBW), RBW in the presence of GMO produces a good and special structure. The substitution of mono and diglycerides (MDG)/polysorbate 80 for GMO causes the structural defect. Ice cream containing RBW/GMO melts more slowly than MDG/polysorbate 80. However, the stability of air bubbles in the gelled oil samples is lower than that in samples with crystalline fat.^[73,94]

Stearic acid or SMS plus polysorbate component gels

Tween or Polysorbate affects SA and SMS oleogel properties. However, though Tween 80 and Tween 20 are both hydrophilic derivatives of sorbitan esters, their behavior is different in SBO-based stearate oleogel. Accordingly, Tween 20 increases the secondary crystallization and crystallization rate, whereas Tween 80 neither shows the secondary crystallization nor increases the crystallization rate. The effect of Tween 80 on the crystallite size and firmness of stearate oleogel is also contrary to the Tween 20; thus, Tween 80 can be used to modulate the thermal and mechanical properties of oleogel, whereas Tween 20 reduces these characteristics. These differences are related to microarchitecture variations. In this regard, polymorph B and postponement of nucleation are induced by Tween 20, and either polymorph B or C and homogenous nucleation are stabilized by Tween

80.^[54,95] Tween 20, in small quantities, improves the stability and lifetime of SMS/cottonseed, soybean, sesame, corn, and olive oil gels as well.^[96]

Polymeric organogelators plus surfactant component gels

EC as a hydrophobic, the only food-grade cellulose derivative polymer, can structure edible oil. The characteristics of EC oleogel could be modified by the type and concentration of surfactant, the concentration, and viscosity of EC, and the oil type.^[17,97] Due to the amphiphilic nature, surfactants can develop the new organized structures via the interaction with the polar area of EC in oleogels and modify the sol-gel transition process. They can also decrease the gelation temperature. The types with the ability to hydrogen bond formation between surfactant head group and EC increase the gel strength considerably at a very low concentration of surfactant (< 0.5%). The influence of surfactant as a plasticizer on the gelation process depends on its compatibility with the polymer backbone, which comprises a smaller head group of surfactant with higher plasticizing effect. Accordingly, glycerol type surfactants such as GMS and GMO are stronger plasticizers than sorbitan esters surfactants (SMS and SMP). According to the mechanical analysis results, GMS-EC oleogel had the highest hardness rate due to the secondary network formation.^[19,74] On the other hand, SMS (Span 60) is the most efficient surfactant on the reduction of EC oleogel syneresis. It also reduces the oleogel elasticity, glass transition temperature (due to action as plasticizer), and average pore size.^[97] Beside these findings, the presence of EC (at a concentration below its critical concentration level for gelation) in MGs (GMS and GMP at different concentrations) organogel also produces stronger gel with slower polymorphic transition, higher OBC, and longer stability than MG organogels without EC.^[75]

Chitin, as a glucose derivative, is a polymer of *N*-acetylglucosamine. It is the second most abundant natural biopolymer after cellulose. Since crude chitin organogel isn't stable, the effect of surfactant addition on polymer degree of aggregation and its alteration to nanocrystals by acid hydrolysis have been investigated. According to the results, the addition of Span-60, phosphatidylcholine (PC) and enzymatically modified PC, as non-ionic, zwitter-ionic and anionic surfactant respectively, to chitin (crude or nanocrystal form) improves the arrangement of polysaccharide molecules; after increasing its flexibility, it subsequently enhances the stability of network. A combination of chitin nanocrystal and surfactant causes uniform and very stable gel with a smoother surface, better organization of strands, and higher elasticity than crude chitin. The largest and lowest gel strengths are obtained by Span 60 and PC respectively. Because of the ability of chitin in the absorption of water, the organogels are not only harder but also more stable after the addition of water up to 25%wt.^[89]

Conclusion

In recent years, more research on mixed system oleogels has been dedicated to the oleogels of sterol/sterol ester or lecithin in combination with the other gelators. There is also a relative lack of research about the application of multi-component supramolecular oleogels in the food industry. In most cases, the capacity of the synergistic interaction of gelators can be used to adapt the gel properties and reduce the concentration of gelators to conform to the official safety standards and reduce the production costs. The future researches should be focused on

understanding the synergistic interactions between the other food-grade gelators to create novel multi-component gels; investigation should be carried on their properties and possible applications in different food products; constructing foam, film and capsule oleocolloids from oleogels to use them as new products and the incorporation of different components such as antioxidant, anti-aging and anti-inflammatory agents by enzymatic catalysis to produce multifunctional gelators. It seems that, beside the clinical tests, practical aspects, and mechanistic studies of multi-component oleogels in the food industry should be investigated to produce more healthfully manufactured food products.

Declaration of interest statement

The authors confirm that there is not known conflict of interest associated with this publication. This article does not contain any studies with human participants or animals performed by any of the authors. Written informed consent was obtained from all study participants.

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