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STRUCTURED OILS IN FOOD SYSTEMS: OLEOGELS AS SUSTAINABLE ALTERNATIVES TO CONVENTIONAL SOLID FATS

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Abstract. Nowadays, there is an increasing emphasis on the quality of dietary fats in food systems, given that the association of high consumption of saturated and trans fats with increased risks of chronic diseases. Thus, the food industry is looking for alternatives capable of reproducing the functional properties of conventional solid fats. Oleogels have emerged as a promising solution, consisting of structured liquid oils immobilized in a three-dimensional network formed by oleogelators such as waxes, monoglycerides, ethylcellulose and biopolymers. The study provides a comprehensive analysis of oleogel systems, focusing on the principles of formation, classification and structuring mechanisms. It highlights the influence of processing parameters, including cooling rate, shear and gelator selection, on the physicochemical and functional properties of oleogels. In addition, the role of different vegetable oils and extraction methods in determining gel stability and performance is discussed. Oleogels demonstrate significant potential in food applications, serving as replacements for saturated fats in products such as bakery products, confectionery, spreads and meat analogues, while improving nutritional quality. Thus, oleogels represent an innovative and sustainable strategy for the development of healthier food products with desired sensory and functional attributes.

Keywords: *fat replacement, food, oleogelator, plant-based oil.*

Rezumat: Astăzi, se pune un accent tot mai mare pe calitatea grăsimilor alimentare din sistemele alimentare, având în vedere asocierea consumului ridicat de grăsimi saturate și trans cu riscuri crescute de boli cronice. Astfel, industria alimentară caută alternative capabile să reproducă proprietățile funcționale ale grăsimilor solide convenționale. Oleogelurile au apărut ca o soluție promițătoare, constând din uleiuri lichide structurate imobilizate într-o rețea tridimensională formată din oleogelatori precum ceruri, monogliceride, etilceluloză și

biopolimeri. Studiul oferă o analiză cuprinzătoare a sistemelor oleogel, concentrându-se pe principiile mecanismelor de formare, clasificare și structurare. Acesta evidențiază influența parametrilor de procesare, inclusiv viteza de răcire, forfecarea și selecția gelatorului, asupra proprietăților fizico-chimice și funcționale ale oleogelurilor. În plus, este discutat rolul diferitelor uleiuri vegetale și al metodelor de extracție în determinarea stabilității și performanței gelului. Oleogelurile demonstrează un potențial semnificativ în aplicațiile alimentare, servind ca înlocuitori pentru grăsimile saturate în produse precum produse de panificație, cofetărie, creme tartinabile și analogi de carne, îmbunătățind în același timp calitatea nutrițională. Astfel, oleogelurile reprezintă o strategie inovatoare și sustenabilă pentru dezvoltarea unor produse alimentare mai sănătoase, cu atribute senzoriale și funcționale dorite.

Cuvinte cheie: *înlocuitor de grăsimi, oleogelator, produs alimentar, ulei vegetal.*

1. Introduction

Dietary fat is a key macronutrient for health and provides more source of energy than carbohydrates or proteins [1]. In processed foods, fat enhances organoleptic characteristics. This kind of fats is generally classified into saturated fatty acids (SFA) and unsaturated fatty acids (UFA) [2]. The major problem with processed animal products is their high fat content, which is dominated by saturated fats and trans fatty acids.

In recent decades, global awareness of the adverse health effects associated with diets rich in saturated and trans fats has increased substantially. Excessive intake of these lipids has been strongly associated with a higher risk of cardiovascular diseases, obesity, type 2 diabetes, and other chronic disorders [3;4]. In response to these concerns, the World Health Organization (WHO) has launched several initiatives aimed at reducing the presence of such fats in food products. Consequently, the food industry has experienced growing pressure to develop innovative alternatives capable of replicating the technological functions of solid fats—such as texture, plasticity, and mouthfeel—while offering an improved nutritional profile [4;5]. Replacing 5% of dietary fats with polyunsaturated fats, such as those derived from vegetable and marine oils, has been reported to reduce the risk of cardiovascular diseases in humans by up to 22–37% [6]. In response to recommendations from nutrition experts and governmental regulations aimed at reducing the use of saturated and trans fats in foods, the field of oil structuring has been extensively investigated over the past decades [7;8].

In this context, oleogels have emerged as a particularly promising and innovative technological approach. Oleogels are semi-solid systems in which a large proportion of liquid oil—typically of vegetable origin and rich in beneficial unsaturated fatty acids—is immobilized within a three-dimensional network generated by a low concentration of structuring agents, commonly referred to as oleogelators [9;10]. Oleogelators, including natural waxes, glycerides, alcohols, ethylcellulose, and phytosterols, facilitate the transformation of vegetable oils into semi-solid systems by creating a structured gel network capable of entrapping liquid oil [2]. The process known as oleogelation enables the conversion of liquid oils into structures exhibiting rheological and textural properties comparable to those of conventional solid fats, such as butter, margarine, without altering the beneficial chemical composition of the oil [9]. This process is considered a nutritionally improved replacement for saturated and trans fats while maintaining the attributes desired by consumers regarding functional and sensory qualities [10]. This unique ability to “solidify” liquid oils opens up extensive opportunities for the reformulation of a wide variety of food

products, including bakery and confectionery items, meat products, and spreads. Furthermore, the versatility of oleogels has attracted considerable attention in other industrial sectors. In the pharmaceutical field, oleogels are being investigated as effective delivery systems for lipophilic drugs, as they can enhance bioavailability and enable controlled release mechanisms [11]. In the cosmetic industry, oleogels provide novel textures and improved stability for creams, lotions, and other personal care formulations, while also serving as carriers for bioactive compounds [12].

Despite the rapid expansion of research in the field of oleogels in recent years, their practical application remains at a relatively early stage of development due to several existing challenges. One of the primary limitations is related to food regulations, which require the use of food-grade structuring agents and often impose restrictions on their permissible concentrations in food products. Although a wide range of oleogels has been developed using different classes of oleogelators, not all of these structuring agents are suitable for food applications [5;13]. Additional constraints include the limited understanding of the interactions between oleogels and other food components, as well as uncertainties regarding their behavior under various processing conditions associated with different food systems. Furthermore, the identification of cost-effective and readily available structuring agents remains an important challenge for the large-scale implementation of oleogels in the food industry.

Nowadays, numerous oleogels are known offering desirable characteristics such as: thermal resistance, texture and improved structure. In addition, foods with oleogels were obtained that demonstrated that they can form textural attributes similar to products made with traditional lipid formulations, that they can improve food nutrition, have high physical and oxidative stability, and a high oil binding capacity [14].

Therefore, the objective of this review is to critically analyze current knowledge on oleogel systems, including the principles of oil structuring, the main classes of oleogelators, their physicochemical and functional characteristics, and their potential applications in food reformulation. In addition, the review highlights the main technological challenges that currently limit the large-scale adoption of oleogels in the food industry.

2. Principles of Oleogel Formation: Classification and Mechanism

The oleogels market is estimated to expand in the coming years at around 6.2% CAGR (compound annual growth rate), supported by increasing demand across the food and beverage, cosmetics and personal care, and coatings sectors. By 2035, the market value is anticipated to reach USD 1.6 billion (Figure 1).

In the food industry, the rising demand for healthier alternatives to saturated and trans fats is a key factor driving the adoption of oleogels. This trend aligns with significant shifts in consumer preferences toward functional, plant-based, and clean-label products. Owing to their favorable textural attributes, mouthfeel, and ability to enhance shelf life, oleogels represent a viable substitute for conventional fats rich in saturated fatty acids. For instance, in confectionery and chocolate applications, oleogels contribute to improved melting behavior and help prevent fat bloom. In plant-based products and processed meat analogues, they effectively replicate the structural and sensory characteristics of animal fat, delivering desirable juiciness and mouthfeel. Furthermore, oleogels can be utilized in the production of vegan cheese, plant-based meat substitutes, margarine, and butter, where they successfully emulate the spreadability, firmness, and sensory properties of traditional fat-based formulations [15].

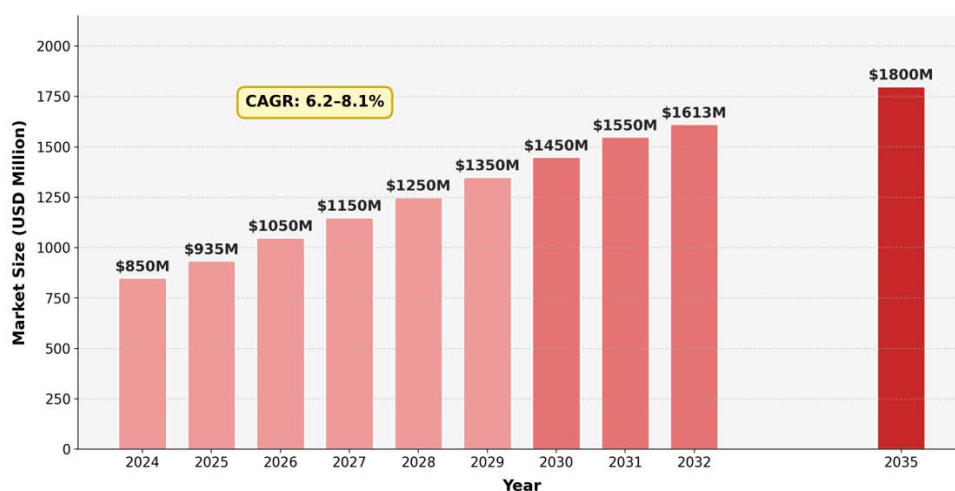


Figure 1. Global oleogels market growth projection from 2024 to 2035.

The industry framework for obtaining oleogels includes several methods categorized into 3 types of methods:

- Direct methods (direct hot method and direct cold method) - provide a process of dispersing waxes, fatty acids, monoglycerides and ethylcellulose, where they are heated until completely dissolved in liquid oil, followed by natural cooling of the mixture to refrigeration temperatures.
- Indirect methods - structured the plant material by adding a polymer to an aqueous solvent or emulsion. The freeze-drying/drying process is used to remove water in order to preserve the structure of the gel network. These methods are appreciated in the case of long-term storage of the formed oleogels because the obtained structure does not show oil removal [16].
- Semi-direct method (capillary suspension) - a small amount of immiscible liquid (usually water) is added to a dispersion of food-grade particles (such as cellulose, soy protein, or starch) in edible oil. This induces the formation of a capillary bridge between the particles, creating a strong three-dimensional network that spans space and immobilizes the oil [17;18].

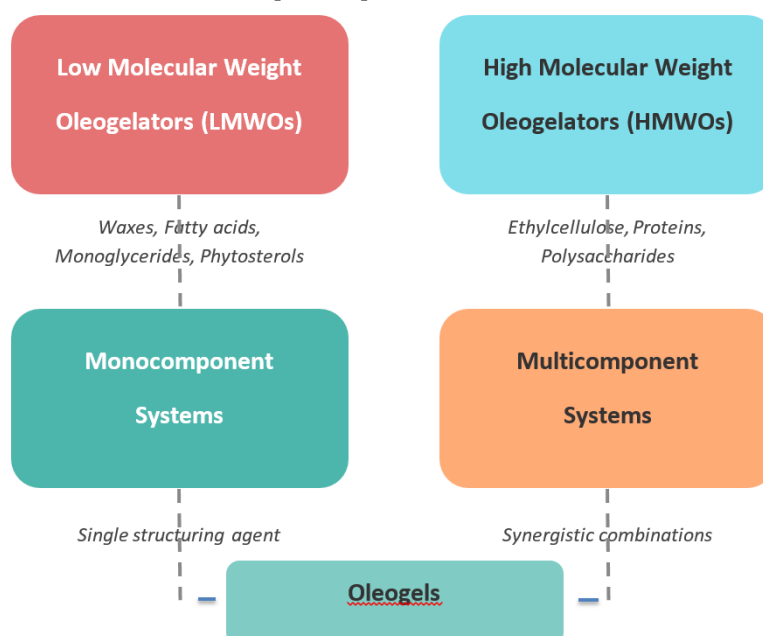


Figure 2. Oleogel classification based on oleogelator type

All of these methods allow the production of oleogels by simpler or more complex techniques. However, there are 3 factors or steps in the methods of obtaining oleogels that are critical, namely:

- Cooling rate: Adequate cooling is essential for the formation of a stable network.
- Shear: Shear applied during gelation can modify the crystal structure.
- Gelator selection: Low molecular weight gelators (LMOG) and high molecular weight gelators (HMOG).

Oleogels are primarily classified according to the chemical nature of the structuring agent, which can be a LMWO, such as waxes or monoglycerides, or a HMWO, such as ethylcellulose, (Figure 2) [19;20]. The formation of the three-dimensional network is a self-assembly process guided by non-covalent physical interactions, being a thermo-reversible phenomenon. For LMWOs, the predominant mechanism is crystallization, where the morphology of the crystals (shape, size, polymorphism) dictates the macroscopic properties of the gel [21]. The efficiency of an oleogelator is defined by its critical gelation concentration (CGC), the minimum value required to form a stable gel [22]. Understanding these fundamental mechanisms is essential for the rational design of oleogels with specific properties.

3. Oleogelators in oleogels production

The selection of the oleogelator is crucial for the functionality of the final product. A wide range of compounds has been explored, each with distinct pro and cons, as summarized in Table 1.

Table 1

Comparative analysis of common oleogelators [6;21]

Oleogelator Type	Structuring Mechanism	Key Advantages	Common Applications
Natural Waxes	Micro-crystal network	Efficient, low cost, GRAS status	Margarine, chocolate, processed meat
Monoglycerides	Lamellar crystal network	Very efficient, fine texture	Confectionery fillings, spreads
Phytosterols/Oryzanol	Nanometric fibrillar network	Health benefits, transparency	Functional foods, pharmaceuticals
Ethylcellulose	Entangled polymer network	Thermal stability, elasticity	Fat replacer in meat, frying
Proteins/Polysaccharides	Particle aggregation	Sustainable sources, clean label	Meat alternatives, vegan foods

*GRAS - Generally Recognized As Safe.

Oleogelators are defined as the main structuring agents responsible for the development of the three-dimensional gel network, whereas “additives” refer to secondary ingredients incorporated to enhance particular properties, such as stability, oxidative

resistance, or emulsifying capacity. Although certain compounds (for instance waxes or proteins) may be included in both groups, their classification is determined by the specific functional role they perform within the formulation. The appropriate selection of biopolymers plays a crucial role in achieving efficient oleogelation. In this regard, proteins and polysaccharides are especially important because of their capacity to form stable gel structures, increase oil-binding ability, and improve the mechanical and sensory characteristics of oleogels [23].

Waxes constitute a diverse group of natural esters that are insoluble in water and particularly effective as moisture barriers. The composition of wax types depends on their source and usually contains different amounts of fatty alcohols, fatty acids and hydrocarbon chains. The ratio between these components is exposed on their gelation behavior in different systems. The advantages of using natural waxes in the formulation of oleogels include good structuring capacity at low concentrations, they are available in sufficient quantities and have an acceptable price [24;25].

The properties of oleogels are influenced by the oil used, oleogelator, structuring agents and the conditions/operations of the technological processing process. These factors determine the degree of structure crystallinity, the gel strength, the oil binding capacity and the behavior of the oleogelator upon melting. Oleogelators exhibit different behaviors during processing, so by selecting and combining the right ingredients, oleogels with customized chemical and physical properties can be formed for subsequent applications in the food industry. Various plant-derived waxes have been extensively studied for their potential use in edible oleogels. Several of these waxes have been classified as GRAS by the US Food and Drug Administration [25;26].

Bryczynski et al. showed that wax esters represent an important component in the formation of the structure of wax-based oleogels, noting that the ester content could range from approximately 15% to about 90% [27].

Beeswax typically exhibited a melting point between 61 and 65 °C, providing moderate thermal stability [28]. It is widely recognized as an effective gelator for structuring edible oils due to its ability to form gels at very low crystalline mass fractions [29].

Carnauba wax showed the highest melting point, generally ranging from 80 to 85 °C, which contributes to its remarkable thermal stability. In addition, it is capable of forming stable gels at concentrations as low as 1.5% [30].

Rice bran wax had a relatively high melting point of approximately 78–82 °C, which ensures excellent thermal stability and favorable crystallization behavior, enabling the formation of stable gel networks [31].

Sunflower wax (SW) exhibited a melting point between 74 and 77 °C, largely due to its high wax ester content, which supports the formation of stable gel structures [32].

Zhang et al. reported that oleogels structured with monoglycerides represent some of the most promising systems, as they are able to crystallize and form strong networks with good elastic properties even at low concentrations (around 3%). In hydrophobic environments, the glycerol head groups of monoglycerides self-assemble into a reverse lamellar phase, while hydrogen bonding between the hydrophilic heads promotes the crystallization of the aliphatic chains. This process leads to oil gelation and contributes to the elasticity of the resulting system [33].

A significant gap remains in the current research regarding the use of phytosterol- γ -oryzanol systems for oleogel preparation. In such complex systems, it is still unclear whether

the gelator and the oil phase can interact synergistically to form a stable crystalline network capable of uniformly distributing water and liquid oil droplets. Furthermore, it remains uncertain whether an excessively dense network could hinder the melting behavior of margarine or whether multi-component crystallization may contribute to achieving the desired textural properties [34]. For instance, Pang et al. investigated the use of sterols combined with beeswax at different concentrations as oleogelators and evaluated their effects on the morphology, oil-binding capacity, texture, color, and stability of the resulting oleogels. The results indicated that, due to the longer chain structure of plant sterol ester, beeswax - plant sterol ester oleogels exhibited enhanced gelation capacity. Moreover, improved oil retention was achieved at lower concentrations, leading to the formation of more robust oleogel structures. In addition, the hardness of beeswax - plant sterol ester oleogels was significantly higher compared with other oleogel systems [35].

Ethylcellulose, a cellulose derivative, is a multifunctional polymer with application in the food industry as a non-toxic additive, as an emulsifier or stabilizer, which can easily replace synthetic additives with the same properties [36;37]. By the direct dispersion method, ethylcellulose can transform oils into oleogels [38], solid and stable, with high nutritional value. However, ethylcellulose can contribute to accelerating the processes of lipid oxidation and reducing the oxidative stability of oleogels, considering the high temperature (125-140 °C) of dissolution of this additive in oil, in the manufacture of oleogels. Nevertheless, ethylcellulose remains an appreciated additive in the production of oleogels, with subsequent use in various branches of the food industry, due to its thermal stability, viscosity control and good binding with various vegetable oils [39].

Qiu et al. obtained ethylcellulose-olive oil oleogels, and the results showed that the oil plays a key role in the gel strength due to its influence on solvent-polymer interactions [40]. Cafuero et al. in their research reported that the quality of oleogels formulated with ethylcellulose can be improved by incorporating additional components, such as phospholipids or mono- and diglycerides. The oleogels obtained by this team based on ethylcellulose and soybean oil, were effectively adjusted in terms of their mechanical and physical properties by modifying the oil phase [41].

While oleogelators such as waxes and phytosterols have been extensively investigated, biopolymers represent relatively novel structuring agents that have been less studied but show considerable potential. These materials may provide promising alternatives due to their enhanced oil-binding capacity, improved thermal stability, and emulsifying properties.

Kirpaci et al. reported that pea protein, although less frequently used in oleogel production, exhibits a high oil-binding capacity and remains effective even at low concentrations, providing improved texture and structural stability [23]. Oleogels based on protein isolates can form strong network structures with minimal oil leakage; however, they also present certain limitations, such as the need for relatively high protein concentrations and potential allergenic effects for specific consumer groups, as highlighted by Sinha et al. [42]. It has also been demonstrated that faba bean protein shows good oil-binding capacity at optimal pH values, although it may present challenges during subsequent technological processing [43]. Similarly, whey protein isolate has exhibited a high oil-binding capacity even at low protein concentrations [44]. In addition, xanthan gum has been reported to provide higher stability, reduced oil loss, and increased gel strength in oleogel systems [45].

3. Plant-based oils used in the production of oleogels

Lipids have applications in foods such as snacks, dairy products, bakery products, and confectionery, due to the need to shape the physical and sensory attributes of these products. For example, solid fats obtained from oils by processes such as hydrogenation, interesterification and fractionation are widely used in a variety of food products. During processing, oils and fats undergo various physicochemical changes that define the product characteristics. An alternative has been considered palm and palm kernel fats, widely used in foods to replace partially hydrogenated fats. However, there are growing concerns about the use of these fats, given their high content of saturated fatty acids and sustainability issues [46]. Thus, the increasing awareness of consumers about the link between diet and health has raised concerns about these fats, especially due to the high levels of saturated and trans fatty acids. As a result, considerable efforts have been made to develop alternative approaches to produce solid fats with a reduced content of saturated fatty acids. [47]. One promising strategy involves structuring liquid oils using oleogelators to create oleogels. In this context, the usefulness of fats and oils in different food products and the modification of fats during processing involve the research of other formulations that can create food products with high nutritional and biological value without the formation of toxic compounds for the health of the consumer [48].

Oils represent about 90% of the composition of an oleogel. In accordance with the oleogel manufacturing process, vegetable oils are chosen according to certain criteria: availability, functionality, low cost, economic importance, nutritional benefits for industrial applications. Vegetable oils with wide application in this field are: soybean oil, olive oil, sunflower oil and sunflower oil with high oleic acid content [49].

The selection of oil for oleogel formulations is determined by the desired properties of the final product. Key considerations include cost, availability, regulatory constraints, and the intended application. In addition, it is essential to evaluate the compatibility and stability of the selected oil with other ingredients and additives to ensure the development of a stable and efficient oleogel system [48]. Some of the most discussed oils in obtaining oleogels are the following.

Soybean oil produced from soybean seeds, that are valued for their agronomic and quality characteristics, making them an important oil crop produced worldwide, providing a nutritionally valuable oil. The US has a soybean production of 8.24 million tons. Brazil contributes 4.28 million tons, Argentina 3.28 million tons, China 3.26 million tons and the EU approximately 2.87 million tons. Soybean oil is composed of 61% polyunsaturated fatty acids, 25% monounsaturated fatty acids and 15% saturated fatty acids [50]. The essential fatty acids are represented in a proportion of 89% by linolenic acid and 11% by α -linolenic acid. Most of the soybean oil (48%) is used in the manufacture of margarine, cooking and salad oils, and mayonnaise.

Rapeseed is characterized as being among the first crops after oil production. **Rapeseed oil** contains approximately 6% saturated fatty acids, is rich in tocopherol, has a lower stability, a property affected by its composition in compounds with high chemical reactivity. It is most often used in the food industry as a vegetable oil or in the manufacture of margarine. [51].

Sunflower oil ranks fourth as the most widely used plant-based oil and, in certain regions, is preferred over soybean, cottonseed, and palm oils. The content of saturated fatty acids does not exceed 10%, in significant quantities it is oleic acid (55-75%) and linoleic acid

(15–35%). It is commonly used for cooking, salad dressings, and margarine production; however, it is less suitable for frying due to its relatively low oxidative stability. Nevertheless, it exhibits good flavor stability [52].

Virgin **olive oil** is obtained by mechanical or other physical processes under specific thermal conditions that do not cause alterations in the oil and is not subjected to other technological treatments specific to the field of obtaining and processing plant-based oils. It is a mixture of triacylglycerols, with some fatty acids, especially palmitic, oleic and linoleic acids. It contains approximately 71% oleic acid [53] and as minor components tocopherols or phenolic compounds with antioxidant properties [54].

Corn oil is extracted from corn kernels, which have a low oil content (3–5%), while the germ contains about 30% of oil. It has a pleasant aroma and about 15% of saturated fatty acids. The oil is characterized by very low α -linolenic acid content and high levels of polyunsaturated fatty acids. Among plant-based oils, corn oil is considered one of the most abundant sources of phytosterols (8300–25,500 ppm) and tocopherols (1130–1830 ppm). Its predominant phytosterol is β -sitosterol (63–70%), while γ -tocopherol (68–89%) represents the major tocopherol fraction. Typically obtained as a by-product of corn starch production, corn oil is widely used in margarine manufacturing, as well as for cooking and salad dressings [55].

Sesame oil is extracted from sesame seeds, that are rich in oil, about 42–56%. It has a good resistant to oxidation process and is known for its potential health benefits. This type of plant-based oil contains about 82% unsaturated fatty acids, with an equal contribution between oleic and linoleic acid. Sesame is also a rich source of minerals and vitamins, including iron, magnesium, copper, calcium, phytosterols, as well as vitamins B1 (thiamine) and E (tocopherols) [56]. Compared to other plant-based oils, sesame oil exhibits superior oxidative stability, making it suitable for use as a cooking oil. [57].

Rice bran oil is a high-quality and versatile edible oil obtained from the outer layer of rice. It is highly valued in the food industry due to its high smoke point (around 230°C), neutral taste, and significant content of antioxidants such as gamma-oryzanol [58]. It is rich in monounsaturated fatty acids (oleic acid, 38.4%) and polyunsaturated fatty acids (linoleic acid, 34.4%), providing a well-balanced fatty acid profile [59]. It is commonly used in high-temperature processes, including frying and baking, as well as in salad dressings, and serves as a stable and healthier alternative to conventional oils in industrial food applications [58].

Linseed oil is extracted from flax seeds and is rich in unsaturated fatty acids, particularly α -linolenic acid (50%). However, its high linolenic acid content makes it highly prone to oxidation, which can quickly lead to undesirable flavors in food products. Therefore, edible linseed oil should be stored under cool, oxygen-free, and light-protected conditions, often with the addition of antioxidants to enhance its stability. This type of oil is most often used in the preparation of salad dressings, margarine, and other food products as a source of omega-3 fatty acids [60].

Pumpkin seed oil exhibits strong antioxidant, anti-inflammatory, and antibacterial properties, making it a promising candidate for various applications in the food industry [61]. It is a good source of biologically active compounds, being considered a “special oil.” α -tocopherol, γ -tocopherol, α -tocotrienol and γ -tocotrienol are the phytochemical components that contribute to the formation of the antioxidant properties specific to pumpkin seed oil [62]. It is not recommended for frying or cooking over high heat because it has a low smoke point of approximately 160°C.

Research in the field has shown that the gelation reaction of different oleogel systems depends on the chemical structure of the oil, mainly on the chain length of the fatty acids and the number of unsaturated fatty acids. Tatel et al. showed that oils with a higher degree of saturation strengthen the oleogel [63]. Also, five types of oils (rapeseed, rice bran, corn, sunflower, and high-oleic sunflower oil), differing in their degrees of saturation, were analyzed to assess how fatty acid profiles affect wax-induced gel formation. The findings indicated that oils with a higher degree of saturation contributed to the formation of stronger oleogel structures [63].

Other oil characteristics, which have a strong effect on the gelation kinetics and crystallization behavior of oleogels, include the viscosity and polarity of the oil. Researchers have observed that weaker gels form in oils with higher polarity due to substantial interactions between the gelator and the solvent [64]. Oils with long carbon chains and higher viscosity produce gels with higher firmness [65].

The effect on the rheological and microstructural characteristics (pore diameter) of oleogels in relation to the type of oil also are important. For example, the results of Zetzel et al. showed that a larger pore diameter (more stable gel) was observed for rapeseed oil, and the pore diameter was reduced with increasing unsaturation of the oil component [66].

The quality, stability, and structure of the oleogel is also influenced by the method of extraction/obtaining the oil from vegetable raw materials. The extraction method influences the fatty acid composition, the presence of impurities, and the degree of oxidation in the liquid oil, all of which directly affect how the oil interacts with the oleogelator and ultimately determine its rheological behavior [66].

The extraction technique governs the fatty acid composition, impurity content, and oxidative status of the liquid oil, which in turn directly influence its interaction with the oleogelator and the resulting rheological characteristics. Oils obtained through different extraction methods contain variable amounts of minor constituents, including phospholipids, free fatty acids, and tocopherols. These components may either impede or promote the development of the oleogelator's crystalline network, thereby affecting the textural attributes and oil-binding capacity [67;68].

Modern industry, as well as the classical one, proposes and uses three methods of extracting oil from vegetable raw materials:

1. Mechanical extraction (press);
2. Solvent extraction
3. Pre-press and solvent method [69].

Mechanical oil extraction can be performed using two main approaches: hot pressing and cold pressing. In hot pressing, the raw material is subjected to elevated temperatures generated during processing, which facilitates the extraction of a substantial proportion of the oil, typically exceeding 75%. In contrast, cold pressing relies on hydraulic systems that operate without significant heat generation, thereby preserving the natural sensory characteristics of the oil. Moreover, the absence of high temperatures minimizes protein denaturation. Although hot pressing generally yields higher extraction efficiency, it often results in reduced oil quality due to thermal degradation [70]. Consequently, cold-pressed oils are considered superior in terms of maintaining their intrinsic properties and being free from chemical alterations, which has contributed to their growing demand. Nevertheless, the efficiency of cold pressing is influenced by several parameters, including applied pressure, screw rotation speed, moisture content of the raw material, and processing temperature. Cold

pressing offers several advantages, including a relatively simple technological process, ease of implementation, and economic benefits due to low capital and operating costs. However, a major limitation of this method is the relatively high amount of residual oil remaining in the press cake, which can reach up to 10–20% according to some reports [71].

Solvent extraction is commonly applied to oilseeds with an oil content below 20%, particularly in soybean processing, where it plays a key role in the oil industry. This method is highly efficient, leaving less than 1% residual oil in the meal after extraction. However, it is relatively time-consuming, as it requires adequate contact between the solvent and the oil within the seed matrix.

Particle size is an important parameter in this process, as reducing it enhances solvent penetration into the plant material and improves extraction yield. Hexane is the most widely used solvent due to its strong affinity for vegetable oils, appropriate boiling point, and relatively low latent heat of evaporation, which supports energy-efficient recovery during distillation. Additionally, hexane is non-corrosive, chemically stable under processing conditions, neutral toward the extracted oil, and economically accessible. Nevertheless, its high flammability necessitates the use of specialized equipment to ensure safe operation [72].

From the perspective of a circular bioeconomy, alternative, more environmentally friendly solvents such as ethanol, isopropanol, and certain hydrocarbons are increasingly being considered. The efficiency of solvent extraction depends on multiple factors, including the composition and moisture content of the raw material, its pretreatment, the type and temperature of the solvent, and the solvent-to-solid ratio [73].

Despite its advantages, this method has several drawbacks, including prolonged processing time, high solvent consumption, potential degradation of heat-sensitive compounds, and the formation of undesirable by-products [69;72].

The oil obtained by this method also needs to be refined. The stages of refining technology include: degumming to remove phospholipids and mucilaginous gums; removal of free fatty acids, metals and chlorophyll by neutralization; washing and drying to remove traces of water and soaps; bleaching to change the color; deodorization to remove volatile substances and carotenoids [74;75].

The **pre-pressing** followed by solvent extraction method is typically applied to oilseeds with an oil content exceeding 25%. In this approach, the initial oil content of the seeds is reduced to approximately 16–20% through mechanical pressing. The resulting press cake is subsequently subjected to solvent extraction under controlled conditions (around 50 °C for approximately 7 hours), allowing the residual oil to be recovered through solvent addition. Using this technique, the residual oil content in the meal can be reduced to below 0.5% [72]. As previously noted, each extraction method presents specific advantages and limitations. Generally, a key drawback of conventional techniques is the lower oil recovery achieved by mechanical pressing compared to solvent extraction, which may negatively impact economic efficiency. Conversely, the reliance on organic solvents in solvent-based methods, including the pre-pressing and solvent extraction approach, raises significant environmental and safety concerns [69].

Gotor and Rhazi in their work presented schematically (Figure 3) the production of sunflower oil. Subsequently, the oil obtained by this extraction method is subjected to several purification steps [76].

Oil refining after solvent extraction includes five main steps to remove minor components: degumming, neutralization, bleaching; winterization and deodorization.

Schematic representation of these stages in the oil processing process can be seen in the form of various simplified diagrams.

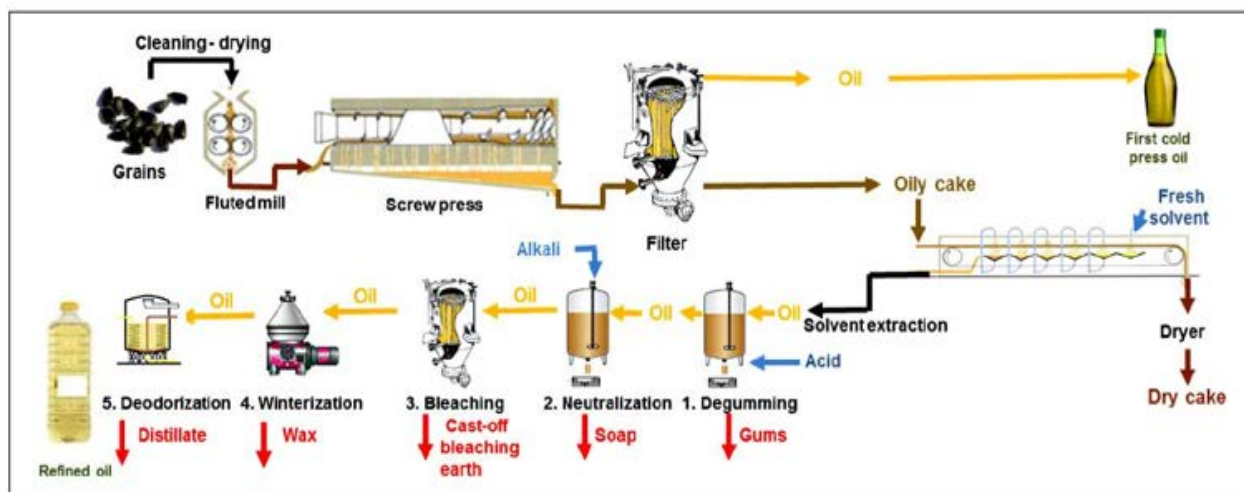


Figure 3. Extracting and refining steps of sunflower oil, [76].

The primary aim of the *degumming* step is to eliminate phospholipids, which are responsible for the dark coloration and undesirable odors of the oil. This process can be carried out using water in the case of hydratable phospholipids, or through acid pretreatment—commonly with phosphoric or citric acid—when non-hydratable phospholipids are present [77]. According to Brevedan et al., degumming can remove approximately 83% of the total phospholipids from solvent-extracted sunflower oil [78].

In addition to phospholipid removal, refining also facilitates the reduction of metal ions. Zufarov et al. reported that acid degumming decreases calcium and magnesium levels by 88% and 90%, respectively [77]. Furthermore, Kreps et al. demonstrated that the combination of degumming and neutralization can reduce free fatty acid content by up to 82% [79]. Degumming can also be carried out through enzymatic treatment. Lamas et al. demonstrated that this approach, when applied to sunflower oil, can achieve substantial reductions in impurities, including up to 97.8% removal of phospholipids, 99.4% reduction in phosphorus content, as well as decreases of 81.2% and 93% in calcium and magnesium levels, respectively [80].

Alkaline *neutralization* of oil or deacidification is an important step in the chemical refining process of edible oil with the aim of neutralizing/removing free fatty acids from the oil, using caustic soda and potassium. These treatments contribute to the transformation of fatty acids into soaps which can subsequently be decanted or separated by centrifugation. Also, this purification step allows to decrease the tocopherol content in the oil, considering that they are unstable in contact with air and alkali. For example, Alpasan et al. showed a decrease of 4.7%, Naz et al. a 16% reduction and Talal et al. a 10% decrease in sunflower oil [81;82;83]. The neutralization process allows the removal of sterols from the oil, in amounts of about 29%. For example, Ruiz-Méndez et al. confirm that a significant amount of eliminated sterols is found in the soap paste obtained from sunflower oil, arguing the influence of the alkali concentration and the temperature used during neutralization on the total content of eliminated sterols [84].

Bleaching earth is a clay-based, mineral-rich material widely used in filtration, adsorption, and decolorization processes. Within the vegetable oil refining sequence, it

represents one of the key steps, where bleaching earth—being the most commonly employed adsorbent—is added to crude edible oil to remove undesirable components. This material is capable of adsorbing a wide range of impurities, including heavy metals, chlorophyll, carotenoids, hydroperoxides, free fatty acids, and various non-glyceride compounds [85]. Bleaching earths are also effective in removing residual soaps, phospholipids, and other polar lipids from the oil [71]. One of the main disadvantages of this step is that the used bleaching earth is usually disposed of in landfills, causing environmental pollution. Therefore, one of the goals of this step should include the processing of this used bleaching earth. From a technological standpoint, bleaching is considered one of the simplest stages in the refining process; however, it is not without drawbacks. During this step, oil losses of up to 2% of the total mass may occur [86]. In practice, bleaching earth is added in quantities exceeding its theoretical adsorption capacity—typically up to 5% (w/w) of the oil—and the mixture is agitated for a defined period. The process is commonly conducted at temperatures above 80°C. Nevertheless, studies indicate that the adsorption efficiency of bleaching earth may decline as the oil temperature increases [74].

Winterization can be generally defined as a physical fractionation process aimed at separating high-melting triglycerides. As a result, the oil is divided into two distinct phases: a viscous liquid fraction and a solid fraction. The solid phase is separated through crystal precipitation followed by filtration, under controlled conditions of temperature, time, and agitation. The crystallization of lipids proceeds through three main stages: supercooling, nucleation, and crystal growth [87]. This method also known as dewaxing, primarily aims to remove long-chain waxes and saturated triglycerides by lowering the oil temperature to approximately 6–8 °C, followed by the separation of the resulting solid fractions. This process targets compounds that crystallize at low temperatures and are responsible for oil turbidity. In the case of sunflower oil, these are predominantly waxes [88;89].

The waxes found in sunflower oil mainly consist of esters formed from long-chain fatty acids and fatty alcohols, typically containing 34 to 50 carbon atoms, with melting points ranging between 70 and 80 °C [90]. Among the refining stages, winterization has the least impact on minor bioactive compounds, particularly tocopherols; for instance, total tocopherol content in sunflower oil decreases by only about 2.4%. Furthermore, Kreps et al. (2014) reported that this process does not significantly affect chlorophyll content [79].

Deodorization represents the final stage of the vegetable oil refining process, designed to enhance oil quality and sensory attributes by removing undesirable odors. This step is both critical and mandatory, with key technological objectives focused on eliminating minor components responsible for off-flavors, including volatile aroma compounds, free and fluorinated fatty acids, and other trace constituents such as tocopherols, sterols, and pesticide residues [91].

During deodorization, the oil is exposed to high temperatures, vacuum or stripping steam, and controlled pressure for a defined period. The process is optimized to maximize the removal of unwanted substances while minimizing the loss of valuable minor components and limiting the formation of trans fatty acids or oxidation products [92]. However, excessively high temperatures can trigger chemical reactions—such as oxidation, cis-trans isomerization, cyclization, and polymerization—that degrade unsaturated fatty acids and generate hazardous contaminants, including 3-monochloropropane-1,2-diol and glycidyl esters [93].

The reduction of minor compounds in deodorized oil, typically less than 20%, partly results from their removal with the injected steam. As a consequence, valuable constituents such as tocopherols, tocotrienols, phytosterols, squalene, and hydrocarbons are concentrated in the deodorization distillate, which represents a significant by-product of edible oil refining. For instance, Naz et al. reported that sunflower oil deodorization distillates contain 13.9–14.2% sterols, 6.5% tocopherols, and 16.5% total hydrocarbons [82].

4. The correlation between the chemical composition of vegetable oils and oleogelation capacity

Vegetable oils are complex lipid systems whose chemical structure and functional properties are profoundly influenced by the nature of their fatty acids and minor bioactive compounds. In the context of oleogel development - semisolid systems obtained through structuring liquid oils - these characteristics are crucial, as they determine the mechanisms of molecular self-assembly and the stability of the three-dimensional networks formed. A comparative analysis of cold-pressed and refined oils highlights significant differences in their oleogelation behavior, rooted in their chemical composition [94].

At the molecular level, vegetable oils are predominantly composed of triacylglycerols (TAGs), esters of glycerol with various fatty acids. For instance, olive oil is rich in oleic acid (MUFA), sunflower oil contains high amounts of linoleic acid (PUFA), and coconut oil is dominated by saturated fatty acids such as lauric acid. These compositional differences directly influence the physical properties of the oils: saturated fatty acids promote ordering and crystallization, whereas polyunsaturated fatty acids increase fluidity and reduce the tendency for molecular organization [95].

In addition to the major TAG fraction, vegetable oils contain a minor fraction composed of phospholipids, phytosterols, tocopherols, and phenolic compounds. This fraction plays a critical role in oleogelation by introducing additional intermolecular interactions, such as hydrogen bonding or dipolar interactions. For example, cold-pressed sesame oil contains lignans and phenolic compounds that can stabilize supramolecular structures, while rapeseed oil contains phytosterols capable of forming fibrillar networks.

The method of oil extraction has a direct impact on the preservation or removal of these components. Cold-pressed oils, such as extra virgin olive oil, flaxseed oil, and pumpkin seed oil, are obtained mechanically at low temperatures, which preserves the minor fraction and the native TAG structure. These oils behave as complex colloidal systems in which amphiphilic compounds (e.g., phospholipids) can act as nucleation agents, facilitating the initiation of crystallization processes. Consequently, during oleogelation, these oils may exhibit an intrinsic capacity for self-structuring, generating heterogeneous three-dimensional networks without the need for large quantities of external oleogelators [96].

In contrast, refined oils, such as refined sunflower oil or soybean oil, undergo intensive purification processes that remove most minor components. The result is an almost ideal lipid system dominated by TAGs, with a reduced level of chemical heterogeneity. In the absence of polar compounds and structuring impurities, nucleation processes are limited, and oleogelation becomes dependent on the addition of external structuring agents, such as beeswax, monoacylglycerols, or purified phytosterols.

The mechanism of oleogelation essentially involves molecular self-assembly and controlled crystallization, mediated by weak intermolecular interactions. Van der Waals forces between hydrocarbon chains play a dominant role, and in the presence of polar

compounds, hydrogen bonding can also occur. In oils rich in saturated fatty acids, such as coconut oil, crystallization occurs rapidly, leading to the formation of rigid networks. In contrast, oils rich in polyunsaturated fatty acids, such as sunflower or flaxseed oils, exhibit higher molecular mobility, which hinders the formation of stable structures [97].

Differences between cold-pressed and refined oils become evident in the formation of oleogels. Cold-pressed oils tend to generate more heterogeneous structures due to the presence of multiple crystallization nuclei and complex molecular interactions. These structures may be less stable over time but offer the advantage of superior nutritional functionality. Refined oils, on the other hand, allow the formation of more uniform and predictable oleogels but require precise control of processing conditions and the addition of specific oleogelators [98].

In conclusion, the oleogelation capacity of vegetable oils results from a complex interplay between chemical composition and processing method. Cold-pressed oils, such as olive, sesame, and flaxseed oils, provide a favorable environment for self-organization due to their richness in minor compounds, whereas refined oils, such as sunflower or soybean oils, require external interventions to develop comparable structures. Understanding these correlations is essential for the design of innovative food products in which structure, stability, and nutritional value must be concurrently optimized.

5. Oleogels in Food Systems: Opportunities and Challenges

Oleogels have been increasingly utilized in the formulation of diverse food products, such as spreads, baked goods, confectionery, dairy, and meat-based items. Beyond their role as substitutes for trans and saturated fats, they exhibit multiple functional properties. Specifically, oleogels can act as delivery systems for lipid-soluble bioactive compounds, contribute to the stabilization of emulsifier-free systems, improve oil retention, and enhance the thermal resistance of food products [97].

However, for all these beneficial aspects of oleogels used in the food industry to be noted in a food product, its compatibility with the food matrix must be ensured, which is currently one of the biggest challenges in the field.

The overall quality of oleogels, as well as the type and quality of the oil employed, significantly influence their characteristics, particularly the sensory attributes.

In addition, processing conditions and subsequent storage, especially exposure to temperature variations, play a crucial role in determining the textural properties of oleogels.

5. Conclusions

This review indicates that the functionality of oleogels is governed by several interrelated factors, including the nature of the oleogelator, the composition of the oil phase, and the applied processing conditions. Collectively, these parameters dictate the structural integrity, oil retention capacity, and overall behavior of oleogels in food systems. Furthermore, the incorporation of plant-based oils and naturally occurring structuring agents contributes to the development of products with a clear label, aligned with sustainability trends. Despite their significant potential, the large-scale industrial application of oleogels remains constrained by regulatory limitations, economic considerations, and an incomplete understanding of their interactions within complex food matrices. Therefore, further research efforts should be directed toward optimizing the formulation, improving oxidative stability, and improving compatibility across diverse food applications. However, oleogels represent an extremely promising strategy for developing healthier and more sustainable fat

alternatives, with substantial potential to advance and redefine contemporary food formulations.

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