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## Review

# Margarines: Historical approach, technological aspects, nutritional profile, and global trends

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## ABSTRACT

Margarines are an expanding market worldwide due to large-scale commercial, lower cost, growth of bakery and confectionery markets, and seasonal independence. The fatty acid composition, solid fat content, consistency, and melting point of the fats used in margarine determine their functional properties. Due to its proven association with increased risk of cardiovascular diseases, the recommendations of the World Health Organization and the enactment of laws in several countries to eliminate industrially produced *trans* fatty acids (TFA) have resulted in the prohibition or progressive reduction in the use of partially hydrogenated fat. However, issues related to high levels of TFA and saturated fatty acids still constitute a challenge in the formulation of this product category. Current trends on margarine production addition of phytosterols, non-lipid components, organogels, and new interesterified fat bases are reviewed. This review aims to present a historical view and the technological evolution of margarines, including their production processes, formulations, and physical and nutritional characteristics, as well as legislation, and main trends.

## 1. Introduction

Margarines are considered water-in-oil emulsions and were introduced as an economically viable alternative to butter (Rajah, 2014). Margarine and low-fat spread consist of an aqueous phase dispersed as fine droplets in liquid oil stabilized within a network of solid fat crystals (Arellano, Norton, & Smith, 2015). US legislation defined margarine as must having at least 80% fat (USDA, 1996). Spreads usually have less than 30%–70% of oil/fat (Patel & Dewettinck, 2016). On the other hand, margarine can be defined as a product with 10%–90% of fat content, depending on the country's legislation is marketed. Since their invention, margarines have undergone a number of changes in relation to their composition and processing, resulting in different products.

The first margarines were prepared with bovine tallow. Over the years, modifications of vegetable oils have been developed and used to replace bovine tallow (Li et al., 2018). For many decades, fats from the partial hydrogenation of vegetable oils were used, which made it possible to obtain solid fats at room temperature (Li, Cobb, Vesper, & Asma, 2019). However, the discovery of health effects related to *trans* fatty acids (TFAs) formed during this hydrogenation process encouraged a number of studies on its replacement (Garcia, Gandra, & Barrera-

Arellano, 2013; Hu, Xu, & Yu, 2017; Li et al., 2018).

In May 2018, the World Health Organization (WHO) launched a set of actions to support governments in removing industrially produced TFAs from the global food supply by 2023. The guidelines recommend the replacement of TFAs in oils and fats, to be achieved through policies and regulations (WHO, 2019). Interesterified fat is currently the main lipid source used in margarines. However, there are still some modifications of fats for margarines that aim to make the product more healthful (Adhikari et al., 2010; Costales-Rodríguez, Gibon, Verhé, & De Greyt, 2009; Li et al., 2018; Renault, 2015).

The promising alternative for making margarines healthier is to replace fats with structured vegetable oils known as organogels (Chaves, Barrera-Arellano, & Ribeiro, 2018). Organogels are a system with a nonpolar liquid organic phase and structuring agents (in low concentrations) promoting the formation of a three-dimensional crystalline network that traps liquid oil, forming a system with solid or semi-solid properties (Hwang, 2020; Rebaka, Rachamalla, Batra, & Subbiah, 2020). Organogels formed by already known structuring agents, whether with a binary, ternary combination or isolated, can be applied in the development of structured emulsions with the potential to reformulate zero-trans and low-sat margarines (Hwang, Singh, Winkler-

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Moser, Bakota, & Liu, 2014; Ögütcü & Yılmaz, 2014; Yılmaz & Ögütcü, 2015). The change in the lipid profile of margarines must be carefully evaluated (Hamley, 2017; Hwang, 2020). The oxidative and physical stability of the reformulated margarine is important for shelf life (Silva et al., 2021). Currently the largest market for margarines and spreads is concentrated in North America (dominated by the United States and Canada), followed by Europe, Asia Pacific, South America, and Africa (Mordor Intelligence, 2020). This market is growing mainly in the Middle East and Africa, with a compound annual growth rate of 2.14% from 2016 to 2025. This growth is linked to the increased consumption of margarines mainly associated with the growth of the bakery and confectionery markets, which use this product as an ingredient. The margarine industry is driven by three main factors: (i) change in the lifestyle of a population in search of more healthful; (ii) a less expensive alternative to butter that provides similar functional properties; (iii) improvements in refinement and methods of lipid modification of vegetable oils that enable the modulation of the functional characteristics of margarines according to need (e.g., increased consistency, aromatization, and presence of essential fatty acids) (Mordor Intelligence, 2020).

The main characteristics of margarines analyzed by consumers are their texture (hardness and spreadability), nutritional composition, and sensory quality (appearance, flavor release, aroma, and color). These properties are determined by the composition and microstructure of the product (i.e., shape, size, and number of crystals; and the strength of the crystalline network formed) (Arellano et al., 2015; Bongers, Almeida, & Hoogland, 2012; Nguyen, Rimaux, Truong, Dewettinck, & Van Bockstaele, 2020; Ögütcü, Arifoğlu, & Yılmaz, 2015). This review aims to present a detailed history of margarines as a reference product and its worldwide reach. First, a discussion of how margarines emerged and became consumed worldwide is presented. The definitions and types of margarines are described, as well as the main functional properties and a traditional manufacturing flowchart. Due to the importance of the lipid phase for the characteristics of margarines, lipid modification methods used and potential alternatives such as organogels are addressed. Legislation to eliminate industrial *trans* fats has affected the whole world, requiring the reformulation of margarines. Thus, we end with new trends and challenges involving this product, taking into account the lifestyle changes of the population and new global legislation.

## 2. Margarines: History and definition

The first margarines were created in 1869 by the French chemist Hippolyte Mege Mouriès (Brown, 1956; Clark, 1986). The discovery came from a contest held by Napoleon III (1808–1873) at a time when France was facing an economic crisis and shortage of some foods, was losing a war, during the Franco-Prussian war of 1870 (Brown, 1956). During this period, there was a great demand for butter, a product prepared with milk fat; however, its production did not meet the market demand (Vaisey-Genser, 2003).

The name “margarine” is derived from the Greek “margaron,” which means “pearl white” (Saillard, 2010). The first company to manufacture margarine was Dutch Jurgens (Clark, 1986). Since then, margarines have undergone several changes in their development in order to improve their functional and sensory characteristics, which has resulted in the wide variety of margarines available on the market. In hard-margarine, these changes include the use of coconut oil, development of modern emulsifiers (monoacylglycerols and diacylglycerols), replacement of coconut oil by cottonseed and soybean oil, addition of synthetic vitamin A, and addition of B-carotene, among others (Aini & Miskandar, 2007; Brown, 1956).

This changes to margarines were based on the premise of removing cholesterol and increasing unsaturated fatty acid content. In addition, it required the transformation of liquid vegetable oils into semi-solid or solid fats at room temperature. This condition was achieved by lipid modification processes such as hydrogenation, which promotes an

increase in the melting point and oxidative stability of the resulting fats compared with that of the starting oil (Kadhun & Shamma, 2017). On the other hand, liquid margarines do not require major changes in their lipid phases, as these consist mainly of vegetable oils rich in unsaturated fat. The main problem involving liquid margarines is the ease of oxidizing at high temperatures (Kirkhus et al., 2015).

In general, table margarine, bakery margarine, puff pastry margarine, vegetable cream, and spreads are considered water-in-oil emulsions in which the lipid concentration varies according to the desired content (between 10 and 90%) (Patel & Dewettinck, 2016; Saillard, 2010). Water, milk or reconstituted milk, emulsifiers, salt, preservatives, vitamins, flavorings, and dyes are additional ingredients (Arellano et al., 2015; Saillard, 2010). In structural terms, margarines are characterized as a crystalline solid with properties of a semi-solid at room temperature. The solid structure of margarine is achieved by a matrix of fat crystal aggregates in which tiny drops of water are entrapped (Arellano et al., 2015).

The types, lipid contents, and properties of margarines are modulated according to the desired application. Margarines with lower lipid content tend to be softer and more spreadable and are intended for application to breads and biscuits, and those with higher lipid content are harder, have higher melting points, and are predominantly used in cooking and baking (Patel, Nicholson, & Marangoni, 2020). The soft type margarine is packed in plastic pots and are spreadable under refrigeration, while the block margarines are packed in tubs and have a firmer consistency. Bakery margarine usually does not require refrigeration, and its formulation is intended to withstand dough working and provide lubrication (Miskandar, Man, Yusoff, & Rahman, 2005).

## 3. General physical properties

The characteristics of margarines result from the ingredients and the production process. Fat defines consistency, plasticity, and melting, and these features are interrelated (Brown, 1956). A good margarine should not suffer oil separation, hardening, sandiness, graininess, water separation, discoloration, or oiliness (Arellano et al., 2015; Miskandar et al., 2005).

### 3.1. Solids profile and melting point

The solid fat content (SFC) that makes up the fat crystals that incorporate liquid oil at a certain temperature is a determining factors in fats, being the property that defines the type of fat to be applied in the product (Goli, Sahri, Kadivar, & Keramat, 2009; Laia, Ghazalia, Cho, & Chong, 2000; Pande, Akoh, & Shewfelt, 2013). The SFC will define characteristics such as general appearance, ease of packaging, organoleptic properties (release of flavor and aroma), spreadability, and oil exudation (Laia et al., 2000), and its determination is carried out using Nuclear Magnetic Resonance (NMR).

Soft or table margarine (which can be used cooled or at ambient temperature) has variable SFC according to the temperature. In the NMR equipment, it is possible to obtain the graph related to the SFC as a function of temperature until the complete melting of the sample. Margarines for use under refrigeration have lower SFC compared with those for use at room temperature (Laia et al., 2000). Between 4 and 10 °C, the spreadability and hardness of margarines under refrigeration are defined, and this should not exceed 32% at 10 °C. Stability is defined between 20 and 22 °C and should not exceed 10% to avoid possible oil exudation. To prevent margarine inducing the feeling of wax in the mouth, the SFC must be less than 3.5%. The thickness and flavor release property are determined at temperatures of 35–37 °C (Laia et al., 2000; Wassell & Young, 2007). Block margarines must have a SFC that makes them spreadable at room temperature without softening at 20 °C. Therefore, they have an SFC of 47–60% at 5 °C, 38–50% at 10 °C, and 19–26% at 20 °C (Sahri & Idris, 2010).

The melting point determines the melting properties of margarines

when consumed in breads and biscuits or in food preparations as an ingredient. At 37 °C, margarines should have low SFC for proper melting at body temperature. The determination of the melting point can also be determined by NMR as being the temperature at which the SFC of margarine is less than 4% (Campos, 2005).

### 3.2. Texture parameters

The main attributes of margarines perceived by the consumer are consistency, hardness, and spreadability (Glibowski, Zarzycki, & Krzepkowska, 2008). These properties can be assessed by subjective methods (sensory analyses) or by instrumental methods (texturometer and penetrometer) (Glibowski et al., 2008). Instrumental methods involve deformations in the structure (Glibowski et al., 2008), and the values obtained can be converted to yield value and thus allow comparisons of the spreadability profile by the scale defined by Haighton (1959) (Fig. 1). The hardness can be expressed in a cone penetrometer. The measurement is quick and the yield value is obtained by the equation below (Esmacilifard & Bahmaei, 2016; Haighton, 1959):

$$C = \frac{K \cdot W}{p^{1.6}}$$

Which,  $C$  is yield value,  $K$  is a constant factor dependent on the angle of the cone,  $W$  is the weight of the cone (grams), and  $p$  is the penetration depth in 0.1 mm. at a penetration during 5 s.

In popular terms, spreadability is the ease with which margarine can be applied in a thin and uniform layer on bread. A spreadable margarine is obtained when there are two phases (liquid and solid oils) in which solid crystals are finely dispersed, and this solid/liquid proportion must melt below body temperature (Miskandar et al., 2005). Adhesiveness is the ease of removing margarine from the mouth or some surface (Bemer, Limbaugh, Cramer, Harper, & Maleky, 2016).

Samples with similar SFC can display different hardness values. This is because the strength of the crystal network depends not only on the amount of solids present but also on the polymorphic behavior and the size of the formed crystal (Glibowski et al., 2008; Laia et al., 2000).

### 3.3. Polymorphism

Polymorphism consists of a crystalline habit in which fats can present themselves. The ability to present different cell structures resulting from various molecular arrangements (Hondoh & Ueno, 2016). In lipids, three specific types of subcells predominate, which refer to primary forms:  $\alpha$ ,  $\beta'$ , and  $\beta$  (Marangoni & Wesdorp, 2012). Polymorph  $\beta'$  is a metastable form with intermediate melting point and is more desirable because it provides a fine arrangement and a large surface area of solid crystals (Marangoni & Wesdorp, 2012). Polymorph  $\beta$ , although the most stable, is not desirable in large quantities because the large crystals of this arrangement will result in a rough and grainy texture (Marangoni & Wesdorp, 2012). Therefore, we seek  $\beta'$  polymorphism in margarines because it promotes plasticity and immobilizes liquid oil (Nguyen et al., 2020).

Palm oil contributes to the  $\beta'$  polymorph and is the best alternative to

partially hydrogenated fats. In addition, there is an influence of external factors associated with the processing and storage of margarines. The  $\beta$  crystal polymorph will produce a margarine that is post-hardened, brittle, grainy, sandy, oily, and greasy (Nguyen et al., 2020).

## 4. Traditional processing of margarines

Traditional margarine production involves stages of transformation of solid or semi-solid fats into a stable emulsified system with spreadable characteristics (Aini & Miskandar, 2007). These main steps will be described, and it is important to highlight that there may be changes according to the fat used, type of margarine, and the desired characteristics in the final product in order to improve its physicochemical, sensory, and nutritional properties. The traditional production of margarines consists of five main stages: preparation of the aqueous phase, preparation of the oil phase, emulsification, crystallization, and packaging/maturation (adapted from Miskandar, Man, Yusoff, & Rahman, 2002) (Fig. 2).

### 4.1. Preparation of the aqueous phase

The aqueous phase is represented by water and water-soluble ingredients such as sodium chloride, antioxidants, acidity regulators, powder milk, and preservatives (Borwankar, Frye, Blaurock, & Sasevich, 1992; Saillard, 2010). These ingredients are added to water to promote complete solubilization. Thickeners and milk proteins can also be added. The aqueous phase is conditioned in a separate tank, and heated to a temperature of approximately 60 °C. Heating of the aqueous phase is necessary so that there is no reduction in temperature during emulsification, which can affect the stability of the emulsion through the early crystallization of ingredients with the highest melting points (Borwankar et al., 1992).

### 4.2. Preparation of the oil phase

The oil phase is composed of oils and/or fats (partially hydrogenated, interesterified, fractionated, or other), plus lipophilic ingredients such as emulsifiers (lecithin, monoacylglycerols, fatty acid polyglycerol esters, sorbitan monostearate, sorbitan tristearate, etc.), colorings ( $\beta$ -carotene, urucum, curcumin, or turmeric extracts), antioxidants, and vitamins (A, D, and E). Its preparation is carried out in a separate tank under agitation and heat above the melting point of the oil or fat (Borwankar et al., 1992).

### 4.3. Emulsification

After heating and solubilization of the ingredients in the aqueous and oil phases, these systems are directed to a single mixing tank for emulsion formation. The emulsification tank features a scraped-surface heating system and agitators and is jacketed for hot- or cold-water flow to allow emulsion temperature control. The emulsion is shaken between 10 and 15 min, and before being transferred to the tube cooler unit, the temperature is increased 2–3 °C above the melting point to

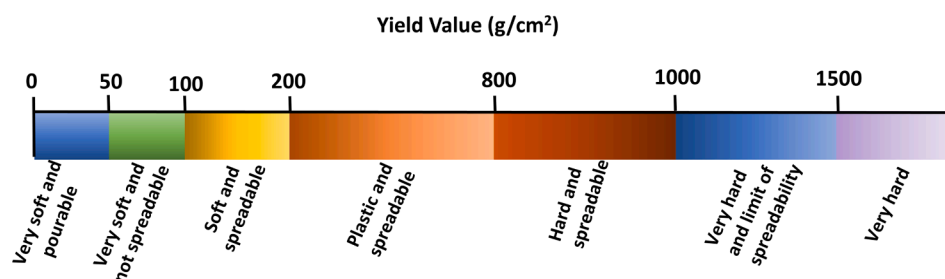


Fig. 1. Classification of margarines according to Yield Value (Haighton, 1959, adapted).

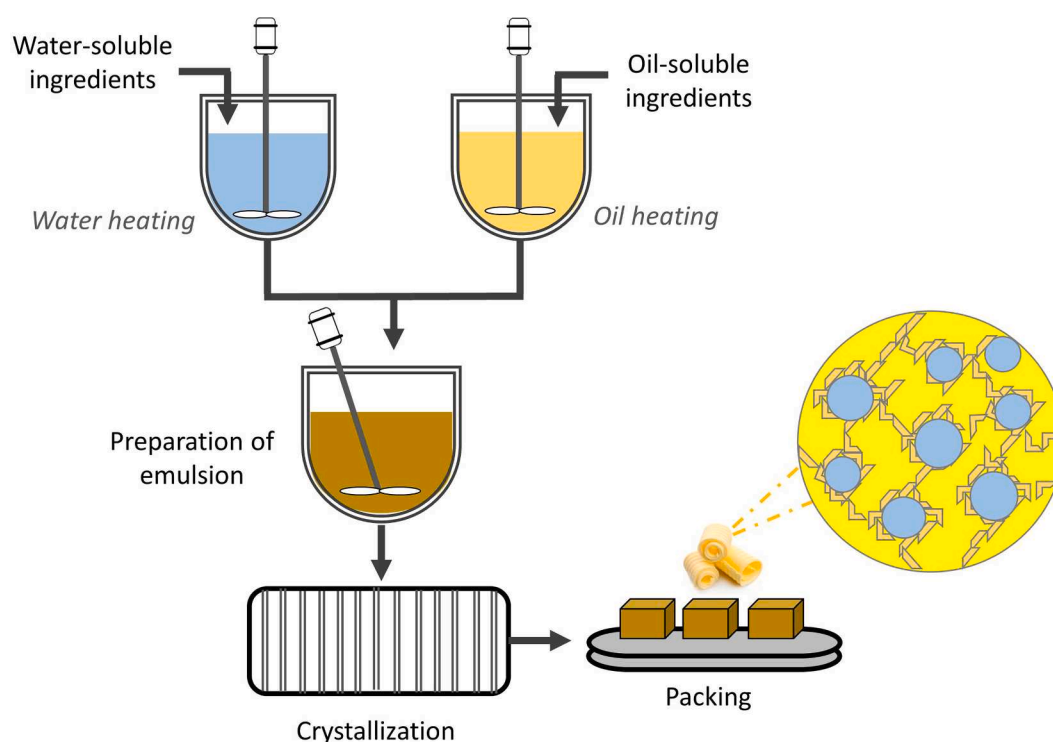


Fig. 2. Conventional margarine production process and schematic representation of its microstructure.

avoid the formation of microscopic crystals (Miskandar et al., 2002). The selected temperature is based on the development of emulsion stability to avoid any pre-crystallization. A suitable emulsion has finely dispersed water droplets in the oily phase (Carr & Vaisey-Genser, 2003).

#### 4.4. Cooling crystallization

After leaving the emulsification tank, the semi-liquid emulsion is directed to the crystallizer. The main step in the processing of margarines is crystallization or plasticization, with formation and maturation of crystals (Borwankar et al., 1992).

The crystallizer consists of scraped-surface heat exchangers with controlled temperatures between 10 and 22 °C. The temperature reduction promotes the formation of many crystalline nuclei from which crystallization proceeds. These crystals are first formed on the walls where the temperature drops rapidly, then are scraped and vigorously amassed (Borwankar et al., 1992). If cooling is slow, a few large crystalline cores are formed. If the cooling is fast, several nuclei of small crystals form (Giacomozzi, Palla, Carrín, & Martini, 2019).

The resulting consistency and plasticity of margarines are consequences of the crystallization behavior determined by their triacylglycerol content. For example, an undesirable crystallization behavior such as the growth of granular crystals affects the softness and spreadability of margarine (Tanaka, Miura, & Yoshioka, 2007). If the crystallization temperature undergoes many fluctuations, the stability of the emulsion may be affected. Critical points of crystallization are considered: (i) insufficient crystallization may result in margarines with low gloss, low creaminess, lumps, low plasticity, and brittle appearance; (ii) excessive crystallization causes margarine to present with excessive gloss and creaminess, oily appearance, low consistency, and migration of oil to the surface (Lima, 2015). When this process occurs in a controlled manner, the fat crystals present the  $\beta'$  polymorphic form, which is suitable for margarines given the feeling of creaminess during consumption and retention of large amounts of liquid oil because of its spherulitic nature (Borwankar et al., 1992).

#### 4.5. Packaging

The packing step is also considered a maturation step (Saillard, 2010). After leaving the crystallizer, the margarine is stored in its own container and brought to stabilization between 5 and 7 °C for approximately 24 h. Margarines should be packed in containers with mechanical, light, and oxygen protection (Carr & Vaisey-Genser, 2003).

##### 4.5.1. Storage stability of the margarines

Margarine with 80% fat typically has a shelf life of 6 to 12 months if refrigerated throughout the distribution and marketing chain. This period is shorter for products low-fat and high humidity and for those that do not contain salt (Vaisey-Genser, 2003). Margarine stability depends mainly on the following factors: composition of the oil phase, emulsifiers, interaction between the ingredients, production process, and crystalline network formatted (Detry et al., 2021).

Margarine stability is associated with changes in its texture, oxidative, and physical properties (Silva et al., 2021). Storage stability is critical from a practical point of view (Zhang, Jacobsen, Pedersen, Christensen, & Adler-Nissen, 2006). Emulsifiers, antioxidants, salt, citric acid, metal ions, or carotenoids are some compounds and ingredients that influence the oxidative stability of margarines (Fruehwirth et al., 2021). The intense oxidation in margarines brings sensory changes as the main problem due to the inadequate taste that is noticeable to the consumer. Antioxidants are usually added to retard the oxidation in margarine as unrestricted activity of free radicals (Han Lyn, Tan, Zawawi, & Nur Hanani, 2021; Nadeem, Imran, Taj, Ajmal, & Junaid, 2017). The oxidative status of margarines can be determined by the peroxide index and sensory analysis (Silva et al., 2018).

The storage temperature of the margarine must be adequately controlled to prevent oxidative reactions from happening. Storage at 5 °C is ideal for this product to maintain its characteristics. When margarine is stored at a temperature of 25 °C, oxidation accelerates (Zhang et al., 2006). Freezing is not indicated because more fat will crystallize, and water droplets can be grouped in the remaining regions of liquid oil, leading to the destabilization of the emulsion after thawing.



(Ghosh & Rousseau, 2011).

During processing, factors such as the size of the crystals and drops are associated with physical stability. Small water droplets, faster crystallization, and immobilization of the aqueous phase promote better stability (Bertoia, Wagner, & Márquez, 2020). The breaking of the emulsion with water or oil separation occurs when the matrix of fine crystals, forming a crystalline network, is not large enough to hold all the water and oil present in the margarines (Detry et al., 2021).

## 5. Lipid modification

Most natural oils have limited application in their unchanged forms, since many are liquid at room temperature. Thus, several methods of lipid modification have emerged: hydrogenation, interesterification, blending, fractionation (Hashempour-Baltork, Torbati, Azadmard-Damirchi, & Savage, 2016), and more recently, use of organogels (Chaves et al., 2018). These processes promote reversible or irreversible changes in vegetable oils, resulting in changes in their physicochemical properties and making them suitable for application. The main food formulated with modified fats has been margarines, the method being chosen according to the technical need, cost, and availability of lipid fractions, regulatory aspects, and nutritional demands of each historical era.

### 5.1. Hydrogenation

Partially hydrogenated fats have excellent functional properties in food, such as crispness in biscuits, aeration in fillings, and texture in margarines, among others. Hydrogenation is the reaction by which the double bonds initially present in the fatty acids are saturated by means of dissolved hydrogen and a catalyst, converting liquid oils into semi-solid or plastic fats (Kadhun & Shamma, 2017). The hydrogenation process increases the melting point and consistency of liquid oils, promotes oxidative stability, and increases technical functionality, allowing for several applications in processed foods (Kadhun & Shamma, 2017). The total hydrogenation process consists of the saturation of the double bonds, getting the hardfats. Hardfats are materials with homogeneous composition, mostly saturated triacylglycerols with a high melting point. These are low-cost materials that have recently been explored by the industry. One of the applications of hardfats is in interesterification, acting as crystallization modulators directing the polymorphic habit of the material (Kadhun & Shamma, 2017; Ribeiro et al., 2015). However, partial hydrogenation leads to the loss of essential fatty acids and causes the formation of geometric isomers (i.e., *cis* isomers are converted into *trans* isomers that are more thermodynamically stable). Studies on lipids and their nutritional properties have highlighted the adverse effects associated with *trans*-fat consumption. These fats are associated with increased cholesterol and risk of coronary heart disease (Brouwer, 2016).

The proposal to eliminate industrially produced TFA was put forward by WHO in 2007, and since then, studies have intensified. However, each country has autonomy in developing legislation to eliminate partially hydrogenated fats from food on the recommendation of WHO (Colón-Ramos, Monge-Rojas, & Campos, 2014; Pan American Health Organization, 2007). Given this scenario, industries have needed to find alternative lipid sources to replace *trans* fats in food. One alternative has been interesterified fats.

### 5.2. Interesterification

Intesterification consists of the chemical or enzymatic reorganization of fatty acids among triacylglycerols; the distribution of fatty acids is altered, but the fatty acid composition remains unchanged (Kadhun & Shamma, 2017). This redistribution of fatty acids promotes changes in the functional properties of lipids, such as improvement in crystallization and fusion behavior, improves consistency, and decreases

the tendency to recrystallize. However, this process has been called into question regarding the formation of triacylglycerol isomers with saturated fatty acids (SFA) in the *sn*-2 position of glycerol (Kadhun & Shamma, 2017; Sivakanthan & Madhujith, 2020). Interesterification is generally used to customize fat with a range of melting points for different food products and to modify crystallization (Adhikari et al., 2010).

The chemical interesterification process uses catalysts such as sodium methoxide and temperatures of 90 °C–110 °C (Sivakanthan & Madhujith, 2020). This reaction promotes the randomization of fatty acids among the three glycerol positions (*sn*-1, *sn*-2, and *sn*-3). The chemical reaction is not specific and therefore development of isomers with SFA at the *sn*-2 glycerol position. In this process, the fatty acid radicals change freely between the triacylglycerol molecules until they reach equilibrium (Kadhun & Shamma, 2017). The chemical process is less expensive, but there is no specificity in the position of the fatty acid in the triacylglycerol (Pande & Akoh, 2013; Zhang, Smith, & Adler-Nissen, 2004).

Enzymatic interesterification is a directed process catalyzed by lipases. Enzymatic interesterification has some advantages over chemistry by producing lipids with specific positions for functional and industrial applications (Zhang, Lee, & Wang, 2020). These enzymes can be non-specific, *sn*-1,3 specific (when they hydrolyze the fatty acids attached to the *sn*-1 and *sn*-3 positions of glycerol), or specific to certain acids fatty acids (when hydrolyzing fatty acids with double bonds in defined positions) (Zhang et al., 2020). The advantages of this process about the chemical stand out: lower temperature, few unit operations, easy separation of enzymes, use of specific enzymes, and more sustainability because it does not involve the use of chemicals (Norizzah, Nur Azimah, & Zaliha, 2018; Zhang et al., 2020).

### 5.3. Fractionation

The fractionation process involves physically separating oils and fats, resulting in products with different melting points. Fractionation consists of a thermomechanical separation process. The oil is cooled until supersaturation to form the crystallization nuclei, progressive growth of the crystalline phase, and separation of the crystalline and liquid phases by vacuum filtration, membranes, or centrifugation (Marikkar, Yanti, Paciulli, & Chiavaro, 2017). The separation of oil fractions is based on the distribution of triacylglycerols between different phases. This depends on the principle of controlled crystallization of triacylglycerols, fractionated in a solid phase (stearin) and a liquid phase (olein) in single fractionation. In double fractionation, the fractions obtained can be fractionated again. Currently, the main fractionated oil applied in food is palm oil (Marikkar et al., 2017; Yilmaz & Ağagündüz, 2020). The objective of fractionation is, in general, a modification of the consistency and behavior of crystallization and fusion. Fractionation is a purely physical process, which minimizes environmental and health concerns (Yilmaz & Ağagündüz, 2020).

### 5.4. Organogels

New lipid modification methods that will produce TFA-free lipid fractions, low SFA content, and high proportion of polyunsaturated fatty acids have been investigated in recent decades (Chaves et al., 2018; Mattice & Marangoni, 2019; Park & Maleky, 2020). Among the alternatives, the structuring of vegetable oils has proved to be feasible in obtaining structured systems with reduced levels of SFA but without chemical or structural modifications of the triacylglycerol molecules. In this approach, liquid oils are structured with or without lipid components to create a structured lipid material (through a three-dimensional network that entraps the oil) capable of providing consistency to food but with a more healthful fatty acid profile. This structuring method is known as organogel or oleogel technology (Chaves et al., 2018). The use of organogels in food products is a attractive alternative, as these

compounds can ensure characteristics such as consistency and plasticity with absence of TFA, and a significant reduction of the SFA content, resulting in products of strong nutritional and technological appeal (Patel et al., 2020; Rogers, Wright, & Marangoni, 2009). Organogels have already been applied to various foods such as cakes, margarines, ice cream, chocolate spreads, meat products, muffins, and cookies (Chaves et al., 2018; Mattice & Marangoni, 2019).

The structuring agents used in the manufacture of organogels are high and low molar mass compounds, which retain liquid oils and form a self-sustaining three-dimensional crystalline network (Rogers, 2009). Structuring agents can be classified into three categories: crystalline particles, self-assembly, and polymeric filaments (Tavernier, Doan, Van der Meeren, Heyman, & Dewettinck, 2018). The choice of structuring agents to be used for the preparation of organogels for food use must be Generally Recognized as Safe (GRAS) (FDA, 2019) and to be able to structure in low concentrations. Organogels applied in foods can be produced with a diversity of structuring agents, the main ones being 12-hydroxystearic acid, ethylcellulose, fatty alcohols, plant sterols, lecithins, waxes, mono- and diacylglycerols, phytosterols/oryzanols, monostearate and sorbitan tristearate, polymers, proteins, and ceramides (Chaves et al., 2018). Organogels with hydrocolloid (locust bean gum, k-carrageenan, xanthan gum, maltodextrin) is also a promising process (Nasirpour-Tabrizi, Azadmard-Damirchi, Hesari, Khakbaz Heshmati, & Savage, 2020).

The use of organogels as substitutes of the lipid phase in margarines has already been reported as potentially feasible, but few studies have accomplished its application (Hwang et al., 2013, 2014; Hwang & Winkler-Moser, 2020; Ögütçü et al., 2015; Ögütçü & Yilmaz, 2014; Silva et al., 2021, 2018; Yilmaz & Ögütçü, 2015, 2014). The organogel suitable for application in margarines should be stable in the emulsion system and not change or destabilize in the presence of the other ingredients in the formulation. The structuring agent 12-hydroxystearic acid has excellent oil structuring properties. However, in the presence of lecithin (an ingredient used in many margarines), this organogel reduces mechanical resistance because 12-hydroxystearic acid forms complexes with lecithin, altering the organogel network (Hwang et al., 2013; Tamura & Ichikawa, 1997).

The main structuring agent in margarines are vegetables waxes (Hwang et al., 2014; Mandu, Barrera-Arellano, Santana, & Fernandes, 2020; Silva et al., 2018; Yilmaz & Ögütçü, 2014). The waxes can crystallize in a network with a high oil binding capacity, and form thermoreversible organogels in very low concentrations (<0.5%) (Tavernier et al., 2018). The criticisms related to wax-based organogels in margarines are residual taste due to the high melting point, flavor of waxes, and emulsion stability (Patel et al., 2020). Hwang et al. (2013) evaluated soybean oil organogels with sunflower wax, rice bran wax, and candelilla wax for application in margarines. The organogel with candelilla wax displayed phase separation after emulsification, rice bran wax showed reduced hardness after incorporation in margarine, and sunflower wax presented the highest hardness (Hwang et al., 2013). Thus, in that study, sunflower wax was the structuring agent with the best characteristics for application in margarines. Margarines with 2%–6% sunflower wax had a higher melting point than conventional margarines. Therefore, the use of waxes provides firmness, melting behavior, and spreadability compatible with the characteristics of commercial margarines. Hwang et al. (2014) developed margarines from sunflower wax organogels and different vegetable oils, with a minimum of 0.3%–1.0% sunflower wax required to obtain firm margarines. Despite these studies showing the potential of waxes in margarines, there are still problems related to interactions when these margarines are applied in food (Hwang, Singh, & Lee, 2016).

Examples of organogels with potential for application in margarines due to their thermal properties, texture, and stability include an organogel with high oleic sunflower oil and myverol (Palla, Giacomozzi, Genovese, & Carrín, 2017), hazelnut oil with beeswax and saturated monoacylglycerols (Yilmaz & Ögütçü, 2014), olive oil with beeswax and

sunflower wax (Yilmaz & Ögütçü, 2015), soybean oil with beeswax (Gao et al., 2021), and soybean oil with zein and  $\beta$ -carotene (Chen et al., 2016). The applications of these organogels have resulted in margarines with characteristics similar to those of commercial products but with a significant reduction in SFA content. The biggest challenge involving these new fats is to find a formulation that presents functional performance, versatility, and desirable sensory properties for application (Mandu et al., 2020).

Mixed organogels (with two or more structuring agents) have been showing potential for application in food, being one of the main tendencies. The rheological behavior and physical stability of some mixed organogels are very similar to edible fats (Rodríguez-Hernández, Pérez-Martínez, Gallegos-Infante, Toro-Vazquez, & Ornelas-Paz, 2021). The main objective in adding more than one structuring agent is to increase the gelation capacity (Hwang, 2020).

Silva et al. (2021) investigate the effect of organogels in low-fat and high-fat margarines during storage. Organogels were developed with candelilla wax, monoacylglycerol, and fully hydrogenated palm oil. The network formed by the organogel protected the oil against oxidation. The amount of water added to the formulations affected the stability, particle size, and texture of the margarines. The combination organogels obtaining a product with reduced SFA levels, zero trans, technological properties similar to conventional margarine, and lower production costs (Silva et al., 2021).

Some properties of organogels are: elastic behavior at low shear rates, non-birefringence, thermoreversibility, thermostability that promotes efficiency in release and medications, transparent, and opaque (Rebaka et al., 2020). The main limitation of the application of organogels in foods is the release of oil (Tavernier et al., 2018) and difficulty in meeting ideal sensory properties for the reformulated product, mainly due to the residual by some structuring agents (Hwang, 2020). Despite this, organogels are an important technological advance in food science due to their versatility, ease of processing, and accessibility (Martins, Vicente, Pastrana, & Cerqueira, 2020).

Studies have been carrying out the replacement of fats in margarines to make them healthier (Park & Maleky, 2020). Stortz, Zetzel, Barbut, Cattaruzza, and Marangoni (2012) evaluated young people who consumed meals with organogels (12-hydroxystearic acid in canola oil) and found that the average levels of serum triacylglycerol were significantly lower compared to young people who ate meals with butter and margarine.

## 6. Nutritional aspects of margarines

The nutritional profile of margarine is defined by the composition of the lipid phase (Dadali & Elmaci, 2019). The preconceptions about fats, are mainly due to the presence of high levels of TFA and SFA. However, these perceptions have changed concomitantly with progressive changes in formulations, and there is a high consumption of margarines in diets worldwide because this foodstuff, in addition to being less expensive, shows higher healthfulness and lower atherogenicity than butter does (Gagliardi, Maranhão, Sousa, Schaefer, & Santos, 2010; Vucic et al., 2015). The atherogenic index (AI) and thrombogenicity index (TI) in food can be used to assess the risk of cardiovascular diseases. Their calculation involves three highly atherogenic fatty acids (lauric, myristic, and palmitic) and unsaturated fatty acids. Overall, the effects of TFA on plasma lipoprotein profiles are at least as unfavorable as those of SFA (Byung, Lumor, & Akoh, 2008). The AI and TI are calculated by the following equations (Vucic et al., 2015):

$$AI = \frac{[C12 : 0 + 4*(C14 : 0) + C16 : 0 + TFA]}{MUFA + (n - 6) + (n - 3)}$$

$$TI = \frac{(C14 : 0 + C16 : 0 + C18) + TFA}{[0.5*MUFA + 0.5*(n - 6) + 3*(n - 3) + (\Sigma^{n-3/n-6})]}$$

Where MUFA are monounsaturated fatty acids, except for TFA.

Miristic acid (C14:0) has a coefficient of 4, as it is considered the most atherogenic of fatty acids. Omega-6 (n-6), least atherogenic, a coefficient of 0.5 was assigned, and Omega-3 (n-3) of 3. TFA are added to the equations because they are also considered to have high atherogenic potential. As an example, margarines in Serbia are reported with AI between 0.23 and 1.67, and TI between 0.44 and 3.04, the higher the TFA content, the higher the margarine TI.

The health effects of TFAs have already been well described, with scientific consensus that their consumption is associated with cardiovascular diseases due to increased low-density lipoprotein (LDL; highly atherogenic), increased total cholesterol levels, and decreased high-density lipoprotein (Brouwer, 2016; Morenga & Montez, 2017). Animal fats also contain TFA, but their presence occurs by a natural mechanism of biohydrogenation. These fatty acids are called vaccenic acids (18:1 t11), positional isomers of animal origin that have been reported for their health-positive biological effects (Poppitt, 2020), and elaidic acid (18:1 t9) a positional isomer produced during partial hydrogenation biggest problem involves industrial TFA, as hydrogenated fat can contain up to 60% TFA and has been widely used in food for many years (Aued-Pimentel & Kus-Yamashita, 2021).

The intake of high levels of TFA and SFA contributes to global epidemics related to metabolic syndrome and cardiovascular diseases (Estadella et al., 2013; Morenga & Montez, 2017). SFAs reportedly promote the increase of LDL, very low-density lipoprotein, cholesterol, and aggregation of blood platelets (Briggs, Petersen, & Kris-Etherton, 2017). In current dietary guidelines, a reduction in SFA consumption in a healthy diet is recommended for the prevention of cardiovascular diseases (Kang, Yang, & Xiao, 2020). According to WHO, the consumption of saturated fats should be less than 10% of total energy consumption, and the intake of TFA less than 1% of total energy consumption (WHO, 2018).

In addition to reducing the consumption of TFA and SFA, increasing polyunsaturated fatty acids (PUFAs) such as omega-3 and omega-6 in the diet is recommended (Hamley, 2017). Depending on the lipid source, margarines can be considered a source of omega-3 and/or omega-6. Vegetable sources of PUFAs include soybean oil, corn oil, canola oil, nuts, flaxseed, and safflower oil (Ohwada, 2018). These fatty acids are important for growth, development, regulation of blood pressure, and plasma lipid levels. However, it is important that the omega-6/omega-3 ratio be ideal (1:2/1) to avoid other problems (Simopoulos & DiNicolantonio, 2016). Margarines usually contain higher levels of omega-6 than omega-3 fatty acids, resulting in an unfavorable ratio (Vucic et al., 2015). The substitution of butter and margarine for unsaturated fats is related to reduced mortality from causes such as cardiovascular diseases, diabetes, cancer, and Alzheimer's disease, mainly associated with the presence of TFA (Zhang et al., 2021).

## 7. Regulatory aspects and legislation

The global margarine market, according to Reports and Data (2019)

is expected to reach \$3.06 billion in 2026 (Reports and Data, 2019). This is due to the growth of the bakery industry in which margarine is fundamental for essential properties such as emulsification, aeration, and lubrication, and growth of market vegan population, who opt for margarine instead of butter (Vucic et al., 2015).

Due to negative reports associated with the consumption of margarine, many countries have improved their understanding about the composition of this food in order to propose new technologies to make it more healthful (Li et al., 2018; Patel, Lecerf, Schenker, & Dewettinck, 2016; Silva et al., 2021). Fig. 3 shows the percentage of countries that have adopted legislation and actions to ban TFA.

Mandatory TFA limits are in place for 2.4 billion people in 28 countries (Argentina, Chile, Colombia, Ecuador, Peru, Saudi Arabia, South Africa, Armenia, Austria, Belarus, Denmark, Hungary, Iceland, Kazakhstan, Kyrgyzstan, Latvia, Norway, Russian Federation, Switzerland, India, Singapore, Thailand, Canada, United States, Romania, Brazil, Slovenia, and Iran) (31% of the global population in 2019) (Li et al., 2019; WHO, 2019). Of these, only 12 countries (Canada, United States, Chile, Latvia, Slovenia, Thailand, Austria, Iceland, Norway, Denmark, Hungary, and South Africa) have best practice policies, covering 540 million people (7% of the global population) (Li et al., 2019; WHO, 2019). Another 24 countries have enacted mandatory TFA limits that will enter into force this year and next (2020 and 2021). In addition, 26 countries (Mexico, Oman, Brazil, Paraguay, Jordan, El Salvador, United Kingdom, Republic of Korea, Brunei Darussalam, Germany, Lithuania, Azerbaijan, Turkey, Bolivia, Spain, Georgia, Philippines, Tunisia, China, France, Belgium, Bulgaria, Israel, Uruguay, Sweden, and Tajikistan) have other complementary measures in place, and 49 countries have a policy, strategy, or national action plan in place that expresses a commitment to reduce TFA in food formulations (WHO, 2019). For the remaining 67 countries, there is no action, or the status is unknown. WHO also recommends that countries develop and implement mandatory TFA limits, share experiences and best practices in eliminating TFA, and consider regional or international networks to improve actions and renew support for the commitment to eliminate industrially produced TFAs by 2023 and achieve the first elimination of a risk factor for non-communicable diseases (WHO, 2019).

Table 1 describes different countries and the contents of TFA, SFA, monounsaturated fatty acids (MUFA), and PUFA present in margarines and spreads. Although most margarines were already suitable for the elimination of TFA, margarines with high *trans* fat levels were still found in Serbia in 2015 (Vucic et al., 2015). In contrast, in United Kingdom in 2013, margarines were already free from TFA (Roe et al., 2013). Countries have been independently adapting to the regulation of TFA content in vegetable oils and margarines (Astrup et al., 2019; WHO, 2019). WHO establishes that the TFA content in vegetable oils and margarines should be less than 2% of the total fat content (Colón-Ramos et al., 2014). In Latin America, Argentina and Chile followed this recommendation to establish the same limit. Puerto Rico and Brazil have ordered a ban on the use of partially hydrogenated fat (Colón-Ramos

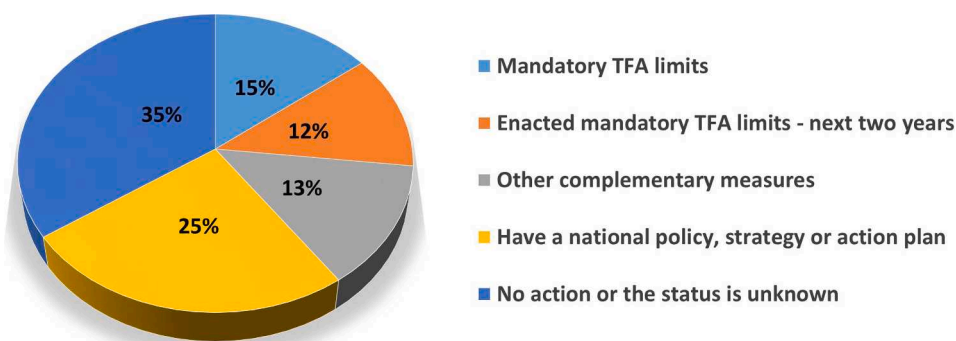


Fig. 3. Percentage of countries in the world and policies implemented to decrease *trans* fatty acids in vegetable oils and fats (Data obtained from WHO, 2019).



**Table 1**

Content of trans, saturated, monounsaturated, and polyunsaturated fatty acids in margarines around the world, in different periods. Values in parentheses represent the average value.

Country	Year	n	SFA (%)	TFA (%)	MUFA (%)	PUFA (%)	Reference
Austria	1996	9	10.1–49.4 (35.4)	0.3–3.7 (1.6)	14.0–43.4 (31.5)	11.04–53.9 (31.4)	Wagner, Auer, and Elmadfa (2000)
German	1996	17	18.74–47.05 (33.03)	0.15–4.88 (1.48)	13.01–34.92 (22.78)	28.89–63.14 (42.70)	Fritsche and Steinhart (1997)
Spain	2000	12	17.5–30.6 (26.8)	0.4–19.2 (5.1)	21–51.1 (31.9)	31.4–52 (41.3)	Larqué, Garaulet, Pérez-Llamas, Zamora, and Tebar (2003)
Czech Republic	2000	20	15.2–54.1 (27.6)	0.1–37.0 (9.7)	42.5–78 (60.1)		Brát and Pokorný (2000)
United States	2002	7	8.5–24.1 (19.5)	15.0–29.3 (19.2)	27.2–34.1 (30.2)	25–34.8 (31.1)	Garsetti et al. (2016)
Portugal	2002	17	23.9–52.6 (32.26)	0.2–8.9 (2.5)	18.4–44.4 (30.1)	10.8–57.4 (32.66)	Torres, Casal, and Oliveira (2002)
Pakistan	2004	10	38.97–53.10 (47.2)	2.45–21.10 (8.34)	21.90–35.80 (28.44)	7–21.50 (13.6)	Anwar, Bhanger, Iqbal, and Sultana (2006)
Turkey	2006	15	11.6–52.1 (33.69)	0.0–39.4 (11.63)	21.0–49.5 (33.23)	1.2–42.8 (21.08)	Karabulut and Turan (2006)
Canada	2007	29	12.4–29.1 (18.5)	0.5–42.9 (11.69)	19.7–59.2 (36.31)	3.6–53.5 (32.09)	Ratnayake et al. (2007)
Costa Rica	2007	50	17.14–37.22 (25.51)	10.83–13.25 (12.12)	24.75–32.03 (28.05)	17.5–46.62 (34.17)	Baylin, Siles, Donovan-Palmer, Fernandez, and Campos (2007)
Pakistan	2008	10	24.2–58.1 (49.75)	22–34.8 (19.99)	5.7–35.4 (15.44)	4.9–37.4 (14.61)	Kandhro et al. (2008)
Greece	2009/10	30	11.26–61.75 (30.81)	0.6–0.97 (0.48)	22.98–60.37 (34.41)	12.09–56.34 (34.28)	Kroustallaki, Tsimpinos, Vardavas, and Kafatos (2011)
Mexico	2011	22 stick	21.66–59.66 (40.92)	0.25–40.65 (8.71)	16.89–32.52 (26.64)	4.41–33.97 (22.04)	Hernández-Martínez, Gallardo-Velázquez, and Osorio-Revilla (2011)
Mexico	2011	20 spreadable	18.07–29.86 (24)	0–20.66 (5.35)	18.3–28.15 (23.05)	24.66–56.51 (45.73)	Hernández-Martínez et al. (2011)
United States	2011	32	17.4–32.4 (24.6)	0.5–23.3 (6.7)	20.8–52.11 (23.6)	26.2–60.6 (45)	Garsetti et al. (2016)
Estonia	2012	12	20.78–37.52 (28.71)	0.04–34.96 (3.48)	25.29–50.52 (43.13)	16.48–39.29 (24.3)	Meremäe, Roasto, Kuusik, Ots, and Henno (2012)
United States	2013	37	10.1–38.9 (26.7)	0.1–21.7 (3.2)	20.8–58.9 (25.9)	12.8–59.3 (44.3)	Garsetti et al. (2016)
Turkey	2013	14	47.1–61.8 (56.15)	1–2.2 (2.2)	21.3–39.3 (30.09)	10.8–18.4 (14.23)	Ergönül (2013)
Serbia	2015	13	22.76–51.17 (33.98)	0.17–28.84 (10.14)	27.28–43.95 (33.03)	8.02–49.29 (21.24)	Vucic et al. (2015)
Slovenia	2018	43	22.6–55.7 (35.3)	0.11–6.37 (0.55)	23.9–51.1 (35.4)	–	Abramovi et al. (2018)

SFA: saturated fatty acids; TFA: trans fatty acids; MUFA: monounsaturated fatty acids; PUFA: polyunsaturated fatty acids; n: number of margarines evaluated.

et al., 2014).

The elimination or reduction of TFA in margarines causes an increase in SFA content, which comes mainly from interesterified fats, in order to promote the functional effects that were provided by TFA (Garsetti, Balentine, Zock, Blom, & Wanders, 2016). WHO recommends that the reformulation of lipid-based foods promote the elimination of TFA, increase PUFA, and decrease SFAs (Astrup et al., 2019). Therefore, there is a recommendation to reduce the intake of SFAs to less than 10% of total energy consumption and replace them with polyunsaturated and monounsaturated fats to reduce the incidence of cardiovascular diseases and related mortality (Chowdhury et al., 2014). It is important to note that not all SFAs are equal in relation to their effects. For example, high plasma concentrations of heptadecanoic acid (17:0) are associated with a reduced risk of coronary heart disease (Astrup et al., 2019). In 2008, the growth of countries with mandatory limits on the TFA content began. To eliminate/reduce the use of partially hydrogenated fats is necessary a fat source that promotes similar characteristics. The alternative found was substitution by saturated fats. As result, the margarines had more saturated fats and consequently with a lower MUFA content. Later, the reduction of saturated fats by healthier sources promoted an increase in MUFA (Kandhro et al., 2008).

## 8. New trends and challenges

The main challenges and trends involving margarines can be categorized as follows: (i) improvement in parameters involving chemical and enzymatic interesterification; (ii) study of structured lipids or new

lipid bases to be interesterified; (iii) use of blends for application; and (iv) use of organogel (Chaves et al., 2018; Costales-Rodríguez et al., 2009; Hwang et al., 2013; Li et al., 2018; Sonwai & Luangsasipong, 2013). These lines currently represent trends in order to produce zero trans margarines, reduced in SFAs and with appropriate functional and sensory properties (Table 2).

Palm oil and its fractions are have excellent properties for food application (Yilmaz & Ağagündüz, 2020). Using a fractionation process, an olein fraction (liquid fraction with high levels of low-melting-point triacylglycerols) and stearin fraction (solid fraction with high levels of high-melting-point triacylglycerols) are obtained (Laia et al., 2000). Its naturally semisolid characteristic at room temperature,  $\beta'$  form crystallization, and the ability to be modified by fractionation, interesterification, hydrogenation, and mixing make palm oil an ideal fat for application in margarines (Ribeiro et al., 2015). In industrial margarines, an average high SFC is sought at 20 °C (Laia et al., 2000). At this temperature, palm oil has 23% solids. Typically, table margarines can be formulated with up to 50% palm oil, with an average of 25%, and when the oil is submitted to interesterification, up to 80% can be used since it promotes physical properties of fats that allow application in margarines. In addition, this oil has a balanced composition of fatty acids (equal concentration of saturated and unsaturated) and contains tocopherols and tocotrienols, providing oxidative stability (Aini & Mis-kandar, 2007).

Palm stearin can be used as a lipid basis in interesterification with liquid oils in order to modify the physical characteristics for the preparation of margarines (Nguyen et al., 2020). The excessive presence of

**Table 2**

Fats developed for application in zero trans and/or reduced saturated margarines.

Methods	Lipid Base	Composition	Reference
Enzymatic interesterification for low-trans margarines	Soybean oil: Fully hydrogenated oil (4:3)	TFA = 0.67%; SFA = 52.6%; UFA = 47.4%	Li et al. (2018)
Hybrid shortenings	Rice bran oil, Sunflower wax, Interesterified palm fat	–	Doan et al. (2017)
Structured lipids – enzymatic	High stearate soybean oil: Palm stearin (2:1)	SFA = 44.1–49.6%; UFA = 57.2–50.9%; TFA = 0	Pande and Akoh (2013)
Organogels	Sunflower wax, rice bran wax, and candelilla wax	–	Hwang et al. (2013)
Blend – Zero-trans Margarines	Virgin coconut oil (VCO): Palm stearin (PS):Palm oil (PO)PS:VCO (40:60 and 30:70) PS:VCO:PO (30:40:30, 20:40:40, 30:50:20, 20:50:30)	TFA = 0–0.16%	Sonwai and Luangsasipong (2013)
Enzymatic interesterification for zero-trans margarines	Pine nut oil: Palm stearin (40:60 and 30:70)	SFA = 40.7 and 47.2%; UFA = 59.3 and 52.8%	Adhikari et al. (2010)
Chemical and enzymatic interesterification for low-trans margarines	Palm stearin: Soybean oil (70:30)	TFA = 0.11%; SFA = 54.8 and 51.5%; UFA = 48.2 and 48.4%	Costales-Rodríguez et al. (2009)
Structured lipids – trans-free margarines	Canola oil: Palm stearin: Palm kernel oil (40:60:0, 40:50:10, 40:40:20, 40:30:30, 50:30:20, and 60:25:15)	SFA = 32.2–52.2%; UFA = 47.8–67.8%	Byung et al. (2008)
Transesterified – enzymatic	Palm stearin: Palm kernel olein (40:60)	–	Laia et al. (2000).
Structured lipids – enzymatic	High-laurate canola oil: Stearic acid (10–80%)	SFA = 60.47%; UFA = 39.53%	Fomuso and Akoh (2001)

TFA: trans fatty acid; SFA: saturated fatty acid; UFA: unsaturated fatty acid.

high melting triacylglycerols, such as palmitic-palmitic-palmitic, leads to the formation of granular crystals in margarines as crystal nuclei. Once nucleation occurs, palmitic-oleic-palmitic (POP), stearic-oleic-stearic, or stearic-stearic-stearic become components of growing crystals. POP triacylglycerols promote the development of undesirable granular crystals in oil-water emulsions, affecting their spreadability and creaminess (Tanaka et al., 2007).

A wide range of fats applied in margarines can be formulated through the interesterification of vegetable oils (Adhikari et al., 2010; Costales-Rodríguez et al., 2009). Currently, most commercial margarines are produced from chemically interesterified fat based on soybean, palm, and cotton oils in mixtures from the original and fully hydrogenated versions.

Pande et al. (2013) evaluated structured lipids obtained from an enzymatic reaction with Novozyme 435 applied in margarines. The resulting product presented a balanced fatty acid profile (with the main fatty acids being palmitic, oleic, stearic, and linoleic), desirable physical properties, and  $\beta'$  polymorphism (Pande et al., 2013). In comparisons of

this margarine (without TFA) with a commercial one (19.1% TFA), the former presented lower spreadability, but there was no difference in terms of flavor. The advantage of the enzymatic process for the production of structured lipids is the use of *sn*-1,3-specific lipases, which maintain the *sn*-2 position of the triacylglycerol intact, keeping the nutritional benefits associated with the intake of fatty acids in this position (Sivakanthan & Madhujith, 2020).

The use of structured lipids may also be accompanied by the enrichment of margarines, such as with conjugated linoleic acid. However, during storage, some changes may arise, such as a reduction in SFC and the transformation of the  $\beta'$  crystal to  $\beta$  (Goli et al., 2009). Another trend involving margarines focuses on making them more healthful through enrichment with components such as plant sterols (phytosterols), which reduce cholesterol; reduce risk for cardiovascular diseases; protect against cervical, breast, and prostate cancer; and modulate the immune system (Raczyk, Bonte, Matthäus, & Rudzińska, 2018; Vezza et al., 2020).

Sometimes, when there is compatibility between raw materials, mixtures can result in excellent bases (Hashempour-Baltork et al., 2016). The mixture of palm stearin and coconut oil (in a 30:70 ratio) presents a trend towards  $\beta'$  crystallization and low SFC at body temperature and does not give a waxy feeling in the mouth. The properties of this mixture allow its application in margarine, which in turn presents with a good consistency and hardness close to that of the commercial one (Sonwai & Luangsasipong, 2013).

Given the current scenario involving nutritional issues with modified oils and fats, margarine has become the target of research to replace partially hydrogenated or interesterified vegetable fat by organogels prepared with different oils and structuring agents (Hwang et al., 2014). Margarines developed with sunflower wax organogel (1%) is an example of the application of this system in the replacement of traditional fats. The maximum amount of palm fat replaceable by organogel has been found to be up to 40% in fats and 25% in margarines. Higher amounts of sunflower wax (2.5%) can reduce up to 40% of saturated fats in hybrid emulsions (Doan, Tavernier, & Danthine, 2017).

Fat mimetics are substances that can mimic some of the organoleptic and physical properties of conventional fat molecules (O'Connor & O'Brien, 2011). The first fat substitute was the Olestra, a mixture of hexa-, hepta- and octa-esters, forming a sucrose polyester. This fat substitute has characteristics very similar to conventional fats, and serves as a non-caloric substitute for dietary fats (Hunt, Zorich, & Thomson, 1998). The new generation of fat mimetics effectively has demonstrated its functionality in numerous applications, since these fall into the clean label category and are very attractive options for the replacement of solid fats. These fats still represent a challenge for application in margarines. The main challenges are producing appropriate sensation in the mouth, firmness, and long-term stability (Patel et al., 2020). However, lowering the fat content causes some textural and organoleptic problems. Other hydrocolloids such as locust bean gum,  $\kappa$ -carrageenan, xanthan gum, and maltodextrin have already had their effects tested on the physical, structural, and sensory properties of low-fat spreads; this new product could be a healthy alternative for spreadable fat and margarine, with low levels of saturated and total fat content (Nasirpour-Tabrizi et al., 2020).

Finally, light margarines are also an expanding market (Mordor Intelligence, 2020). Light margarines are those with less than 60% fat and are mainly used in breads and biscuits. They have higher levels of unsaturated fatty acids or functional enrichment nutrients. These products present good cardiovascular effects and contribute to the maintenance or reduction of blood cholesterol (Saillard, 2010).

Therefore, the challenges involving margarines focus mainly on finding technical fats that replace industrial *trans* fats without having a sharp increase in the content of SFA or altering functional and sensory properties. At the same time, these lipid systems must have a similar cost to those currently used so that the product does not incur significant cost changes. Margarines and spreads with lower lipid content represent the

greatest difficulty in obtaining these characteristics due to problems with emulsion stability.

## 9. Conclusion

Despite nutritional issues involving the consumption of margarines, such as TFA still present in formulations and the high levels of SFA, this market is expanding worldwide. Legislation involving these fatty acids is becoming more restrictive, forcing industries to modify the fats available. The emergence of lipid modification methods is one of the greatest innovations in the processing of margarines. With these, changes in the stages involving the processing and properties of these fats have arisen. Special attention needs to be paid to the evaluation of the influence of parameters such as solid content, melting point, polymorphism, and fat consistency in margarines during storage. Apart from these issues, countries have invested in research margarines more healthful, as consumers worry about consuming food that causes adverse health effects. The availability of a low-cost product such as margarine, would promote an increase in the consumption of this product which presents ideal functional properties in accordance with nutritional issues (SFA reduction, enrichment with phytosterols, lipid content reduction, and recourse of bioactive compounds) and that promote the increase in the consumption of this product. Furthermore, its functionality as an ingredient, especially in the bakery and confectionery industry, has caused the expansion of this market. Therefore, more research should be performed in order to reformulate margarines to achieve ideal physical, sensory, and nutritional properties for the current market.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## References

- Abramovi, H., Vidrih, R., Zlati, E., Kokalj, D., Schreiner, M., Katja, Ž., ... Pravst, I. (2018). Journal of Food Composition and Analysis Trans fatty acids in margarines and shortenings in the food supply in Slovenia. *Journal of Food Composition and Analysis*, 74(January), 53–61. <https://doi.org/10.1016/j.jfca.2018.08.007>.
- Adhikari, P., Zhu, X. M., Gautam, A., Shin, J. A., Hu, J. N., Lee, J. H., ... Lee, K. T. (2010). Scaled-up production of zero-trans margarine fat using pine nut oil and palm stearin. *Food Chemistry*, 119(4), 1332–1338. <https://doi.org/10.1016/j.foodchem.2009.09.009>.
- Aini, I. N., & Miskandar, M. S. (2007). Utilization of palm oil and palm products in shortenings and margarines. *European Journal of Lipid Science and Technology*, 109 (4), 422–432. <https://doi.org/10.1002/ejlt.200600232>.
- Anwar, F., Bhangar, M. I., Iqbal, S., & Sultan, B. (2006). Fatty acid composition of different margarines and butters from Pakistan with special emphasis on trans unsaturated contents. *Journal of Food Quality*, 29(1), 87–96. <https://doi.org/10.1111/j.1745-4557.2006.00058.x>.
- Arellano, M., Norton, I. T., & Smith, P. (2015). Specialty oils and fats in margarines and low-fat spreads. In *Specialty Oils and Fats in Food and Nutrition: Properties, Processing and Applications* (pp. 242–270). Elsevier Inc.. <https://doi.org/10.1016/B978-1-78242-376-8.00010-7>.
- Astrup, A., Bertram, H. C. S., Bonjour, J. P., De Groot, L. C. P., De Oliveira Otto, M. C., Feeney, E. L., ... Soedamah-Muthu, S. S. (2019). WHO draft guidelines on dietary saturated and trans fatty acids: Time for a new approach? *The BMJ*, 366(July), 1–6. <https://doi.org/10.1136/bmj.l4137>.
- Aued-Pimentel, S., & Kus-Yamashita, M. M. (2021). Analysis of the fat profile of industrialized food in Brazil with emphasis on trans-fatty acids. *Journal of Food Composition and Analysis*, 97, Article 103799. <https://doi.org/10.1016/j.jfca.2020.103799>.
- Baylin, A., Siles, X., Donovan-Palmer, A., Fernandez, X., & Campos, H. (2007). Fatty acid composition of Costa Rican foods including trans fatty acid content. *Journal of Food Composition and Analysis*, 20(3–4), 182–192. <https://doi.org/10.1016/j.jfca.2006.01.004>.
- Bemer, H. L., Limbaugh, M., Cramer, E. D., Harper, W. J., & Maleky, F. (2016). Vegetable organogels incorporation in cream cheese products. *Food Research International*, 85, 67–75. <https://doi.org/10.1016/j.foodres.2016.04.016>.
- Bertoia, L., Wagner, J. R., & Márquez, A. L. (2020). Margarine-like emulsions prepared with coconut and palm oils: Analysis of microstructure and freeze–thaw stability by differential scanning calorimetry. *JAOCs, Journal of the American Oil Chemists' Society*, 97(10), 1071–1081. <https://doi.org/10.1002/aocs.12399>.
- Bongers, P., Almeida, C., & Hoogland, H. (2012). Dynamic modelling of margarine manufacturing. *Computer Aided Chemical Engineering*, 30(June), 532–536. <https://doi.org/10.1016/B978-0-444-59519-5.50107-6>.
- Borwankar, R. P., Frye, L. A., Blaurock, A. E., & Sasevich, F. J. (1992). Rheological characterization of melting of margarines and tablespreads. *Journal of Food Engineering*, 16(1–2), 55–74. [https://doi.org/10.1016/0260-8774\(92\)90020-7](https://doi.org/10.1016/0260-8774(92)90020-7).
- Brát, J., & Pokorný, J. (2000). Fatty acid composition of margarines and cooking fats available on the Czech market. *Journal of Food Composition and Analysis*, 13(4), 337–343. <https://doi.org/10.1006/jfca.1999.0877>.
- Briggs, M., Petersen, K., & Kris-Etherton, P. (2017). Saturated fatty acids and cardiovascular disease: Replacements for saturated fat to reduce cardiovascular risk. *Healthcare*, 5(2), 29. <https://doi.org/10.3390/healthcare5020029>.
- Brouwer, I. (2016). *Effects of trans-fatty acid intake on blood lipids and lipoproteins: a systematic review and meta-regression analysis* (p. 84). Geneva: World Health Organization.
- Brown, L. C. (1956). Margarine production. *Journal of the American Oil Chemists' Society*, 33(10), 506–512. <https://doi.org/10.1007/BF02612309>.
- Byung, H. K., Lumor, S. E., & Akoh, C. C. (2008). Trans-free margarines prepared with canola oil/palm stearin/palm kernel oil-based structured lipids. *Journal of Agricultural and Food Chemistry*, 56(17), 8195–8205. <https://doi.org/10.1021/jf801412v>.
- Campos, R. (2005). Experimental methodology. In A. G. Marangoni (Ed.), *Fat Crystal Networks* (pp. 267–349). New York: Marcel Dekker.
- Carr, R. A., & Vaisey-Genser, M. (2003). Methods of Manufacture. *Encyclopedia of Food Sciences and Nutrition*, 1990, 3709–3714. <https://doi.org/10.1016/B0-12-227055-X/00739-2>.
- Chaves, K. F., Barrera-Arellano, D., & Ribeiro, A. P. B. (2018). Potential application of lipid organogels for food industry. *Food Research International*, 105, 863–872. <https://doi.org/10.1016/j.foodres.2017.12.020>.
- Chen, X. W., Fu, S. Y., Hou, J. J., Guo, J., Wang, J. M., & Yang, X. Q. (2016). Zein based oil-in-glycerol emulsions enriched with  $\beta$ -carotene as margarine alternatives. *Food Chemistry*, 211, 836–844. <https://doi.org/10.1016/j.foodchem.2016.05.133>.
- Chowdhury, R., Warnakula, S., Kunutsor, S., Crowe, F., Ward, H. A., Johnson, L., ... Di Angelantonio, E. (2014). Association of dietary, circulating, and supplement fatty acids with coronary risk: A systematic review and meta-analysis. *Annals of Internal Medicine*, 160(6), 398–406. <https://doi.org/10.7326/M13-1788>.
- Clark, P. (1986). The Marketing of Margarine. *European Journal of Marketing*, 20(5), 52–65. <https://doi.org/10.1108/EUM000000004647>.
- Colón-Ramos, U., Monge-Rojas, R., & Campos, H. (2014). Impact of WHO recommendations to eliminate industrial trans-fatty acids from the food supply in Latin America and the Caribbean. *Health Policy and Planning*, 29(5), 529–541. <https://doi.org/10.1093/heapol/czt034>.
- Costales-Rodríguez, R., Gibon, V., Verhé, R., & De Greyt, W. (2009). Chemical and enzymatic interesterification of a blend of palm stearin: Soybean oil for low trans-margarine formulation. *Journal of the American Oil Chemists' Society*, 86(7), 681–697. <https://doi.org/10.1007/s11746-009-1395-2>.
- Dadali, C., & Elmaci, Y. (2019). Characterization of volatile release and sensory properties of model margarines by changing fat and emulsifier content. *European Journal of Lipid Science and Technology*, 121(6), 1–8. <https://doi.org/10.1002/ejlt.201900003>.
- Detry, R., Van Hoed, V., Sterckx, J., Deledicque, C., Sato, K., Blecker, C., & Danthine, S. (2021). Physicochemical properties of palm oil-based puff pastry model margarines related to their baking performance in long-term storage. *European Journal of Lipid Science and Technology*, 123(1), 1–12. <https://doi.org/10.1002/ejlt.202000155>.
- Doan, C. D., Tavernier, I., & Danthine, S. (2017). Physical compatibility between wax esters and triglycerides in hybrid shortenings and margarines prepared in rice bran oil. *Journal of the Science of Food and Agriculture*, 98, 1042–1051. <https://doi.org/10.1002/jsfa.8553>.
- Ergönül, P. G. (2013). Solid fat contents and instrumental textural attributes of margarines sold in Turkish market. *Quality Assurance and Safety of Crops & Foods*, 5 (2), 157–161. <https://doi.org/10.3920/qas2012.0122>.
- Esmailifard, N., & Bahmaei, M. (2016). Comparison of physicochemical characteristics of some margarines and butters in Iranian market during storage. *Journal of Pharmaceutical and Health Sciences*, 4(3), 181–192.
- Estadella, D., Nascimento, C. M., Oyama, L. M., Ribeiro, E. B., Dámaso, A. R., & Piano, A. (2013). Lipotoxicity: Effects of dietary saturated and trans fatty acids. *Mediators of Inflammation*, Article 137579. <https://doi.org/10.1155/2013/137579>.
- Fomuso, L. B., & Akoh, C. C. (2001). Enzymatic modification of high-laurate canola to produce margarine fat. *Journal of Agricultural and Food Chemistry*, 49(9), 4482–4487. <https://doi.org/10.1021/jf010444u>.
- Fritzsche, J., & Steinhart, H. (1997). Trans fatty acid content in German margarines. *Fett/Lipid*, 99(6), 214–217. <https://doi.org/10.1002/lipi.19970990604>.



- Fruehwirth, S., Egger, S., Kurzbach, D., Windisch, J., Jirsa, F., Flecker, T., & Pignitter, M. (2021). Ingredient-dependent extent of lipid oxidation in margarine. *Antioxidants*, 10(1), 1–17. <https://doi.org/10.3390/antiox10010105>.
- Gagliardi, A. C. M., Maranhão, R. C., Sousa, H. P. D., Schaefer, E. J., & Santos, R. D. (2010). Effects of margarines and butter consumption on lipid profiles, inflammation markers and lipid transfer to HDL particles in free-living subjects with the metabolic syndrome. *European Journal of Clinical Nutrition*, 64(10), 1141–1149. <https://doi.org/10.1038/ejcn.2010.122>.
- Gao, Y., Lei, Y., Wu, Y., Liang, H., Li, J., Pei, Y., ... Liu, S. (2021). Beeswax: A potential self-emulsifying agent for the construction of thermal-sensitive food W/O emulsion. *Food Chemistry*, 349, Article 129203. <https://doi.org/10.1016/j.foodchem.2021.129203>.
- García, R. K. A., Gandra, K. M., & Barrera-Arellano, D. (2013). Development of a zero trans margarine from soybean-based interesterified fats formulated using artificial neural networks. *Grasas y Aceites*, 64(5), 521–530. <https://doi.org/10.3989/gya.049113>.
- Garsetti, M., Balentine, D. A., Zock, P. L., Blom, W. A. M., & Wanders, A. J. (2016). Fat composition of vegetable oil spreads and margarines in the USA in 2013: A national marketplace analysis. *International Journal of Food Sciences and Nutrition*, 67(4), 372–382. <https://doi.org/10.3109/09637486.2016.1161012>.
- Ghosh, S., & Rousseau, D. (2011). Fat crystals and water-in-oil emulsion stability. *Current Opinion in Colloid and Interface Science*, 16(5), 421–431. <https://doi.org/10.1016/j.cocis.2011.06.006>.
- Giacomozzi, A. S., Palla, C. A., Carrín, M. E., & Martini, S. (2019). Physical properties of monoglycerides oleogels modified by concentration, cooling rate, and high-intensity ultrasound. *Journal of Food Science*, 84(9), 2549–2561. <https://doi.org/10.1111/1750-3841.14762>.
- Glibowski, P., Zarzycki, P., & Krzepkowska, M. (2008). The rheological and instrumental textural properties of selected table fats. *International Journal of Food Properties*, 11(3), 678–686. <https://doi.org/10.1080/10942910701622599>.
- Goli, S. A. H., Sahri, M. M., Kadiivar, M., & Keramat, J. (2009). The production of an experimental table margarine enriched with conjugated linoleic acid (CLA): Physical properties. *Journal of the American Oil Chemists' Society*, 86(5), 453–458. <https://doi.org/10.1007/s11746-009-1362-y>.
- Haighton, A. J. (1959). The measurement of the hardness of margarine and fats with cone penetrometers. *Journal of the American Oil Chemists Society*, 36(8), 345–348. <https://doi.org/10.1007/BF02640051>.
- Hamley, S. (2017). The effect of replacing saturated fat with mostly n-6 polyunsaturated fat on coronary heart disease: A meta-analysis of randomised controlled trials. *Nutrition Journal*, 16(1), 1–16. <https://doi.org/10.1186/s12937-017-0254-5>.
- Han Lyn, F., Tan, C. P., Zawawi, R. M., & Nur Hanani, Z. A. (2021). Physicochemical properties of chitosan/ graphene oxide composite films and their effects on storage stability of palm-oil based margarine. *Food Hydrocolloids*, 117(November 2020), 106707. <https://doi.org/10.1016/j.foodhyd.2021.106707>.
- Hashempour-Baltork, F., Torbati, M., Azadmard-Damirchi, S., & Savage, G. P. (2016). Vegetable oil blending: A review of physicochemical, nutritional and health effects. *Trends in Food Science and Technology*, 57, 52–58. <https://doi.org/10.1016/j.tifs.2016.09.007>.
- Hernández-Martínez, M., Gallardo-Velázquez, T., & Osorio-Revilla, G. (2011). Fatty acid profile including trans fatty acid content of margarines marketed in Mexico. *JAOCs, Journal of the American Oil Chemists' Society*, 88(10), 1485–1495. <https://doi.org/10.1007/s11746-011-1815-y>.
- Hondoh, H., & Ueno, S. (2016). Polymorphism of edible fat crystals. *Progress in Crystal Growth and Characterization of Materials*, 62(2), 398–399. <https://doi.org/10.1016/j.pcrysgrow.2016.04.021>.
- Hu, P., Xu, X., & Yu, L. L. (2017). Effect of fatty acid chain length on the crystallization behavior of trans-free margarine basestocks during storage. *Journal of Oleo Science*, 66(4), 353–362. <https://doi.org/10.5650/jos.ess16210>.
- Hunt, R., Zorich, N. L., & Thomson, A. B. (1998). Overview of olestra: a new fat substitute. *Canadian Journal of Gastroenterology and Hepatology*, 12(3), 193–197. <https://doi.org/10.1155/1998/389685>. PMID: 9582544.
- Hwang, H. S. (2020). A critical review on structures, health effects, oxidative stability, and sensory properties of oleogels. *Biocatalysis and Agricultural Biotechnology*, 26, Article 101657. <https://doi.org/10.1016/j.bcab.2020.101657>.
- Hwang, H. S., Singh, M., Bakota, E. L., Winkler-Moser, J. K., Kim, S., & Liu, S. X. (2013). Margarine from organogels of plant wax and soybean oil. *Journal of the American Oil Chemists' Society*, 90(11), 1705–1712. <https://doi.org/10.1007/s11746-013-2315-z>.
- Hwang, H. S., Singh, M., & Lee, S. (2016). Properties of cookies made with natural wax-vegetable oil organogels. *Journal of Food Science*, 81(5), C1045–C1054. <https://doi.org/10.1111/1750-3841.13279>.
- Hwang, H. S., Singh, M., Winkler-Moser, J. K., Bakota, E. L., & Liu, S. X. (2014). Preparation of margarines from organogels of sunflower wax and vegetable oils. *Journal of Food Science*, 79(10), C1926–C1932. <https://doi.org/10.1111/1750-3841.12596>.
- Hwang, H. S., & Winkler-Moser, J. K. (2020). Properties of margarines prepared from soybean oil oleogels with mixtures of candelilla wax and beeswax. *Journal of Food Science*, 85(10), 3293–3302. <https://doi.org/10.1111/1750-3841.15444>.
- Kadhun, A. A. H., & Shamma, M. N. (2017). Edible lipids modification processes: A review. *Critical Reviews in Food Science and Nutrition*, 57(1), 48–58. <https://doi.org/10.1080/10408398.2013.848834>.
- Kandhro, A., Sherazi, S. T. H., Mahesar, S. A., Bhangar, M. I., Younis Talpur, M., & Rauf, A. (2008). GC-MS quantification of fatty acid profile including trans FA in the locally manufactured margarines of Pakistan. *Food Chemistry*, 109(1), 207–211. <https://doi.org/10.1016/j.foodchem.2007.12.029>.
- Kang, Z. Q., Yang, Y., & Xiao, B. (2020). Dietary saturated fat intake and risk of stroke: Systematic review and dose-response meta-analysis of prospective cohort studies. *Nutrition, Metabolism and Cardiovascular Diseases*, 30(2), 179–189. <https://doi.org/10.1016/j.numecd.2019.09.028>.
- Karabulut, I., & Turan, S. (2006). Some properties of margarines and shortenings marketed in Turkey. *Journal of Food Composition and Analysis*, 19(1), 55–58. <https://doi.org/10.1016/j.jfca.2004.06.016>.
- Kirkhus, B., Lamblait, A., Storø, I., Vogt, G., Olsen, E., Lundby, F., & Standal, H. (2015). The role of water in protection against thermal deterioration of liquid margarine. *JAOCs, Journal of the American Oil Chemists' Society*, 92(2), 215–223. <https://doi.org/10.1007/s11746-014-2589-9>.
- Kroustallaki, P., Tsimpinos, G., Vardavas, C. I., & Kafatos, A. (2011). Fatty acid composition of Greek margarines and their change in fatty acid content over the past decades. *International Journal of Food Sciences and Nutrition*, 62(7), 685–691. <https://doi.org/10.3109/09637486.2011.568473>.
- Laia, O. M., Ghazalia, H. M., Cho, F., & Chong, C. L. (2000). Physical and textural properties of an experimental table margarine prepared from lipase-catalysed transesterified palm stearin: Palm kernel olein mixture during storage. *Food Chemistry*, 71(2), 173–179. [https://doi.org/10.1016/S0308-8146\(00\)00084-4](https://doi.org/10.1016/S0308-8146(00)00084-4).
- Larqué, E., Garaulet, M., Pérez-Llamas, F., Zamora, S., & Tebar, F. J. (2003). Fatty acid composition and nutritional relevance of most widely consumed margarines in Spain. *Grasas y Aceites*, 54(1), 65–70. <https://doi.org/10.3989/gya.2003.v54.i1.279>.
- Li, C., Cobb, L. K., Vesper, H. W., & Asma, S. (2019). Global surveillance of trans-fatty acids. *Preventing Chronic Disease*, 16(10), 1–5. <https://doi.org/10.5888/pcd16.190121>.
- Li, Y., Zhao, J., Xie, X., Zhang, Z., Zhang, N., & Wang, Y. (2018). A low trans margarine fat analog to beef tallow for healthier formulations: Optimization of enzymatic interesterification using soybean oil and fully hydrogenated palm oil. *Food Chemistry*, 255(January), 405–413. <https://doi.org/10.1016/j.foodchem.2018.02.086>.
- Lima, J. F. F. L. (2015). *Aplicação e definição de metodologias para melhoria contínua no processo de produção na área das margarinas*. Dissertação: Universidade Nova de Lisboa.
- Mandu, C. C., Barrera-Arellano, D., Santana, M. H. A., & Fernandes, G. D. (2020). Waxes used as structuring agents for food organogels: A review. *Grasas y Aceites*, 71(1), 1–13. <https://doi.org/10.3989/gya.1169182>.
- Marangoni, A. G., & Wesdorp, L. H. (2012). *Structure and properties of fat crystal networks* (2nd ed.). CRC Press.
- Marikkar, N., Yanti, N., Paciulli, M., & Chiavaro, E. (2017). Chemical composition and thermal behaviour of tropical fat fractions from solvent-assisted process: A review. *Italian Journal of Food Science*, 29(1), 19–37. <https://doi.org/10.14674/1120-1770/ijfs.v566>.
- Martins, A. J., Vicente, A. A., Pastrana, L. M., & Cerqueira, M. A. (2020). Oleogels for development of health-promoting food products. *Food Science and Human Wellness*, 9(1), 31–39. <https://doi.org/10.1016/j.fshw.2019.12.001>.
- Mattice, K. D., & Marangoni, A. G. (2019). Oleogels in Food. In *Encyclopedia of Food Chemistry* (pp. 255–260). Elsevier. <https://doi.org/10.1016/B978-0-08-100596-5.21662-4>.
- Meremäe, K., Roasto, M., Kuusik, S., Ots, M., & Henno, M. (2012). Trans fatty acid contents in selected dietary fats in the Estonian market. *Journal of Food Science*, 77(8). <https://doi.org/10.1111/j.1750-3841.2012.02829.x>.
- Miskandar, M. S., Man, Y. B. C., Yusoff, M. S. A., & Rahman, R. A. (2002). Effect of emulsion temperature on physical properties of palm oil-based margarine. *Journal of the American Oil Chemists' Society*, 79(12). <https://doi.org/10.1007/s11746-002-0621-4>.
- Miskandar, M. S., Man, Y. B. C., Yusoff, M. S. A., & Rahman, R. A. (2005). Quality of margarine: Fats selection and processing parameters. *Asia Pacific Journal of Clinical Nutrition*, 14(4), 387–395.
- Mordor Intelligence (2020). Global margarine market – Growth, trends, and forecasts (2020–2025). <https://www.mordorintelligence.com/industry-reports/margarine-market>.
- Morenga, L. Te, & Montez, J. M. (2017). Health effects of saturated and trans-fatty acid intake in children and adolescents: Systematic review and meta-analysis. *PLoS ONE*, 12(11), Article e0186672. <https://doi.org/10.1371/journal.pone.0186672>.
- Nadeem, M., Imran, M., Taj, I., Ajmal, M., & Junaid, M. (2017). Omega-3 fatty acids, phenolic compounds and antioxidant characteristics of chia oil supplemented margarine. *Lipids in Health and Disease*, 16(1), 1–12. <https://doi.org/10.1186/s12944-017-0490-x>.
- Nasirpour-Tabrizi, P., Azadmard-Damirchi, S., Hesari, J., Khakbaz Heshmati, M., & Savage, G. P. (2020). Production of a spreadable emulsion gel using flaxseed oil in a matrix of hydrocolloids. *Journal of Food Processing and Preservation*, 44(8), 1–10. <https://doi.org/10.1111/jfpp.14588>.
- Nguyen, V., Rimaux, T., Truong, V., Dewettinck, K., & Van Bockstaele, F. (2020). Granular crystals in palm oil based shortening/margarine: A review. *Crystal Growth and Design*, 20(2), 1363–1372. <https://doi.org/10.1021/acs.cgd.9b01191>.
- Norizzah, A. R., Nur Azimah, K., & Zaliha, O. (2018). Influence of enzymatic and chemical interesterification on crystallization properties of refined, bleached and deodorised (RBD) palm oil and RBD palm kernel oil blends. *Food Research International*, 106, 982–991. <https://doi.org/10.1016/j.foodres.2018.02.001>.
- O'Connor, T. P., & O'Brien, N. M. (2011). Butter and other milk fat products – Fat Replacers. *Encyclopedia of Dairy Sciences*, 528–532.
- Ögütçü, M., Arifoğlu, N., & Yılmaz, E. (2015). Preparation and characterization of virgin olive oil-beeswax oleogel emulsion products. *Journal of the American Oil Chemists' Society*, 92(4), 459–471. <https://doi.org/10.1007/s11746-015-2615-6>.
- Ögütçü, M., & Yılmaz, E. (2014). Oleogels of virgin olive oil with carnauba wax and monoglyceride as spreadable products. *Grasas y Aceites*, 65(3). <https://doi.org/10.3989/gya.0349141>.



- Ohwada, T. (2018). Polyunsaturated fatty acids (PUFAs) in human health. In *Polyunsaturated Fatty Acids (PUFAs): Food Sources, Health Effects and Significance in Biochemistry* (pp. 41–70).
- Palla, C., Giacomozzi, A., Genovese, D. B., & Carrín, M. E. (2017). Multi-objective optimization of high oleic sunflower oil and monoglycerides oleogels: Searching for rheological and textural properties similar to margarine. *Food Structure*, 12, 1–14. <https://doi.org/10.1016/j.foodstr.2017.02.005>.
- Pan American Health Organization (2007). PAHO/WHO task force trans fats free Americas: conclusions and recommendations (Issue April 26–27). <http://www1.paho.org/English/DD/PIN/pr070607.htm>.
- Pande, G., & Akoh, C. C. (2013). Enzymatic synthesis of trans-free structured margarine fat analogs with high stearate soybean oil and palm stearin and their characterization. *LWT – Food Science and Technology*, 50(1), 232–239. <https://doi.org/10.1016/j.lwt.2012.05.027>.
- Pande, G., Akoh, C. C., & Shewfelt, R. L. (2013). Utilization of enzymatically interesterified cottonseed oil and palm stearin-based structured lipid in the production of trans-free margarine. *Biocatalysis and Agricultural Biotechnology*, 2(1), 76–84. <https://doi.org/10.1016/j.bcab.2012.08.005>.
- Park, C., & Maleky, F. (2020). A critical review of the last 10 years of oleogels in food. *Frontiers in Sustainable Food Systems*, 4, 1–8. <https://doi.org/10.3389/fsufs.2020.00139>.
- Patel, A. R., & Dewettinck, K. (2016). Edible oil structuring: An overview and recent updates. *Food Function*, 7, 20–29. <https://doi.org/10.1039/C5FO01006C>.
- Patel, A. R., Lecerf, J. M., Schenker, S., & Dewettinck, K. (2016). The contribution of modern margarine and fat spreads to dietary fat intake. *Comprehensive Reviews in Food Science and Food Safety*, 15(3), 633–645. <https://doi.org/10.1111/1541-4337.12198>.
- Patel, A. R., Nicholson, R. A., & Marangoni, A. G. (2020). Applications of fat mimetics for the replacement of saturated and hydrogenated fat in food products. *Current Opinion in Food Science*, 33, 61–68. <https://doi.org/10.1016/j.cofs.2019.12.008>.
- Poppitt, S. D. (2020). Cow's milk and dairy consumption: Is there now consensus for cardiometabolic health? *Frontiers in Nutrition*, 7(December), 1–22. <https://doi.org/10.3389/fnut.2020.574725>.
- Raczyk, M., Bonte, A., Matthäus, B., & Rudzińska, M. (2018). Impact of added phytosteryl/phytostanyl fatty acid esters on chemical parameters of margarines upon heating and pan-frying. *European Journal of Lipid Science and Technology*, 120(2), 1–11. <https://doi.org/10.1002/ejlt.201700281>.
- Rajiah, K. K. (2014). Spreadable products. In *Fats in Food Technology* (2nd ed., pp. 213–252). Wiley.
- Ratnayake, W. M. N., Gagnon, C., Dumais, L., Lillycrop, W., Wong, L., Meleta, M., & Calway, P. (2007). Trans Fatty acid content of Canadian margarines prior to mandatory trans fat labelling. *Journal of the American Oil Chemists' Society*, 84(9), 817–825. <https://doi.org/10.1007/s11746-007-1112-y>.
- Rebaka, V. P., Rachamalla, A. K., Batra, S., & Subbiah, N. (2020). In *State of the Art and New perspectives in oleogels and applications* (pp. 151–182). Cham: Springer. [https://doi.org/10.1007/978-3-030-42284-4\\_6](https://doi.org/10.1007/978-3-030-42284-4_6).
- Renault, A. (2015). Margarines with linseed oil: Nutritional interests, specificities and development. *OC – Oilseeds and Fats*, 22(6), 4–7. <https://doi.org/10.1051/ocl/2015024>.
- Reports and Data (2019). Industrial Margarine Market To Reach USD 3.06 Billion By 2026. Retrieved from <https://www.globenewswire.com/news-release/2019/11/19/1949677/0/en/Industrial-Margarine-Market-To-Reach-USD-3-06-Billion-By-2026-Reports-And-Data.html>.
- Ribeiro, A. P. B., Masuchi, M. H., Miyasaki, E. K., Domingues, M. A. F., Stroppa, V. L. Z., de Oliveira, G. M., & Kieckbusch, T. G. (2015). Crystallization modifiers in lipid systems. *Journal of Food Science and Technology*, 52(7), 3925–3946. <https://doi.org/10.1007/s13197-014-1587-0>.
- Rodríguez-Hernández, A. K., Pérez-Martínez, J. D., Gallegos-Infante, J. A., Toro-Vazquez, J. F., & Ornelas-Paz, J. J. (2021). Rheological properties of ethyl cellulose-monoglyceride-candelilla wax oleogel vis-a-vis edible shortenings. *Carbohydrate Polymers*, 252(October 2020). <https://doi.org/10.1016/j.carbpol.2020.117171>.
- Roe, M., Pinchen, H., Church, S., Elahi, S., Walker, M., Farron-Wilson, M., ... Finglas, P. (2013). Trans fatty acids in a range of UK processed foods. *Food Chemistry*, 140(3), 427–431. <https://doi.org/10.1016/j.foodchem.2012.08.067>.
- Rogers, M. A. (2009). Novel structuring strategies for unsaturated fats – Meeting the zero-trans, zero-saturated fat challenge: A review. *Food Research International*, 42(7), 747–753. <https://doi.org/10.1016/j.foodres.2009.02.024>.
- Rogers, M. A., Wright, A. J., & Marangoni, A. G. (2009). Oil organogels: the fat of the future? *Soft Matter*, 5(8), 1594–1596. <https://doi.org/10.1039/b822008p>.
- Sahri, M. M., & Idris, N. A. (2010). Palm stearin as low trans hard stock for margarine. *Sains Malaysiana*, 39(5), 821–827.
- Saillard, M. (2010). Margarines et matières grasses tartinables. *Cahiers de Nutrition et de Dietétique*, 45(5), 274–280. <https://doi.org/10.1016/j.cnd.2010.08.001>.
- Silva, T. L. T., Chaves, K. F., Fernandes, G. D., Rodrigues, J. B., Bolini, H. M. A., & Arellano, D. B. (2018). Sensory and technological evaluation of margarines with reduced saturated fatty acid contents using oleogel technology. *Journal of the American Oil Chemists' Society*, 95(June), 673–685. <https://doi.org/10.1002/aocs.12074>.
- Silva, T. J., Fernandes, G. D., Bernardinelli, O. D., Silva, E. C. da R., Barrera-Arellano, D., & Ribeiro, A. P. B. (2021). Organogels in low-fat and high-fat margarine: A study of physical properties and shelf life. *Food Research International*, 140(October 2020), 110036. <https://doi.org/10.1016/j.foodres.2020.110036>.
- Simopoulos, A. P., & DiNicolantonio, J. J. (2016). The importance of a balanced  $\omega$ -6 to  $\omega$ -3 ratio in the prevention and management of obesity. *Open Heart*, 3(2), 1–6. <https://doi.org/10.1136/openhrt-2015-000385>.
- Sivakanthan, S., & Madhujith, T. (2020). Current trends in applications of enzymatic interesterification of fats and oils: A review. *LWT – Food Science and Technology*, 132, Article 109880. <https://doi.org/10.1016/j.lwt.2020.109880>.
- Sonwai, S., & Luangsasipong, V. (2013). Production of zero-trans margarines from blends of virgin coconut oil, palm stearin and palm oil. *Food Science and Technology Research*, 19(3), 425–437. <https://doi.org/10.3136/fstr.19.425>.
- Stortz, T. A., Zetzl, A. K., Barbut, S., Cattaruzza, A., & Marangoni, A. G. (2012). Edible oleogels in food products to help maximize health benefits and improve nutritional profiles. *Lipid Technology*, 24(7), 151–154. <https://doi.org/10.1002/lite.201200205>.
- Tamura, T., & Ichikawa, M. (1997). Effect of lecithin on organogel formation of 12-hydroxystearic acid. *Journal of the American Oil Chemists' Society*, 74(5), 491–495. <https://doi.org/10.1007/s11746-997-0170-5>.
- Tanaka, L., Miura, S., & Yoshioka, T. (2007). Formation of granular crystals in margarine with excess amount of palm oil. *Journal of the American Oil Chemists' Society*, 84(5), 421–426. <https://doi.org/10.1007/s11746-007-1064-2>.
- Tavernier, I., Doan, C. D., Van der Meer, P., Heyman, B., & Dewettinck, K. (2018). The Potential of Waxes to Alter the Microstructural Properties of Emulsion-Templated Oleogels. *European Journal of Lipid Science and Technology*, 120(3), 1–13. <https://doi.org/10.1002/ejlt.201700393>.
- Torres, D., Casal, S., & Oliveira, M. B. P. (2002). Fatty acid composition of Portuguese spreadable fats with emphasis on trans isomers. *European Food Research and Technology*, 214(2), 108–111. <https://doi.org/10.1007/s00217-001-0418-5>.
- USDA. (1996). *USDA Specifications for vegetable oil margarine*. United States Department of Agriculture.
- Vaisey-Genser, M. (2003). Types and Properties. *Encyclopedia of Food Sciences and Nutrition*, 3704–3709.
- Veza, T., Canet, F., de Marañón, A. M., Bañuls, C., Rocha, M., & Víctor, V. M. (2020). Phytosterols: Nutritional health players in the management of obesity and its related disorders. *Antioxidants*, 9(12), 1–20. <https://doi.org/10.3390/antiox9121266>.
- Vucic, V., Arsic, A., Petrovic, S., Milanovic, S., Gurinovic, M., & Glibetic, M. (2015). Trans fatty acid content in Serbian margarines: Urgent need for legislative changes and consumer information. *Food Chemistry*, 185, 437–440. <https://doi.org/10.1016/j.foodchem.2015.04.018>.
- Wagner, K. H., Auer, E., & Elmadfa, I. (2000). Content of trans fatty acids in margarines, plant oils, fried products and chocolate spreads in Austria. *European Food Research and Technology*, 210(4), 237–241. <https://doi.org/10.1007/s002179900080>.
- Wassell, P., & Young, N. W. G. (2007). Food applications of trans fatty acid substitutes. *International Journal of Food Science and Technology*, 42(5), 503–517. <https://doi.org/10.1111/j.1365-2621.2007.01571.x>.
- WHO. (2018). *Healthy diet*. World Health Organization.
- WHO. (2019). *Countdown to 2023: WHO report on global trans fat elimination 2019*. Geneva: World Health Organization.
- Yilmaz, B., & Ağaçgözü, D. (2020). Fractionated palm oils: Emerging roles in the food industry and possible cardiovascular effects. *Critical Reviews in Food Science and Nutrition*, 1–10. <https://doi.org/10.1080/10408398.2020.1869694>.
- Yilmaz, E., & Öğütcü, M. (2014). Comparative Analysis of Olive Oil Organogels Containing Beeswax and Sunflower Wax with Breakfast Margarine. *Journal of Food Science*, 79(9), E1732–E1738. <https://doi.org/10.1111/1750-3841.12561>.
- Yilmaz, E., & Öğütcü, M. (2015). Oleogels as spreadable fat and butter alternatives: Sensory description and consumer perception. *The Royal Society of Chemistry*, 5, 50259–50267. <https://doi.org/10.1039/C5RA06689A>.
- Zhang, H., Jacobsen, C., Pedersen, L. S., Christensen, M. W., & Adler-Nissen, J. (2006). Storage stability of margarines produced from enzymatically interesterified fats compared to those prepared by conventional methods – Chemical properties. *European Journal of Lipid Science and Technology*, 108(3), 227–238. <https://doi.org/10.1002/ejlt.200500305>.
- Zhang, Z., Lee, W. J., & Wang, Y. (2020). Evaluation of enzymatic interesterification in structured triacylglycerols preparation: A concise review and prospect. *Critical Reviews in Food Science and Nutrition*, 1–15. <https://doi.org/10.1080/10408398.2020.1793725>.
- Zhang, H., Smith, P., & Adler-Nissen, J. (2004). Effects of degree of enzymatic interesterification on the physical properties of margarine fats: Solid fat content, crystallization behavior, crystal morphology, and crystal network. *Journal of Agricultural and Food Chemistry*, 52(14), 4423–4431. <https://doi.org/10.1021/jf035022u>.
- Zhang, Y., Zhuang, P., Wu, F., He, W., Mao, L., Jia, W., ... Jiao, J. (2021). Cooking oil/fat consumption and deaths from cardiometabolic diseases and other causes: Prospective analysis of 521,120 individuals. *BMC Medicine*, 19(1), 1–14. <https://doi.org/10.1186/s12916-021-01961-2>.