Received: 7 September 2019 | Revised: 4 December 2019 | Accepted: 3 January 2020

DOI: 10.1111/1541-4337.12537

COMPREHENSIVE REVIEWS IN FOOD SCIENCE AND FOOD SAFETY

Comprehensive REVIEWS in Food Science and Food Safety WILEY

Synthesis, physicochemical properties, and health aspects of structured lipids: A review

Yalong Guo[1] | **Zhixiang Cai**[1] | **Yanping Xie**[1] | **Aiqin Ma**[2] | **Hongbin Zhang**[1] | **Pingfan Rao**[3] | **Qiang Wang**[4]

[1]Department of Polymer Science and Engineering, School of Chemistry and Chemical Engineering, Advanced Rheology Institute, Shanghai Key Laboratory of Electrical Insulation and Thermal Aging, Shanghai Jiao Tong University, Shanghai, P. R. China

[2]Shanghai Jiao Tong University Affiliated Sixth People's Hospital South Campus, Shanghai, P. R. China

[3]Food Nutrition Sciences Centre, Zhejiang Gongshang University, Hangzhou, P. R. China

[4]Institute of Food Science and Technology, Chinese Academy of Agricultural Sciences, Beijing, P. R. China

Correspondence
Hongbin Zhang, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, No 800 Dongchuan Road, Shanghai 200240, P. R. China.
Email: hbzhang@sjtu.edu.cn

Funding information
National Key Research and Development Plan, Grant/Award Number: 2016YFD0400200

Abstract

Structured lipids (SLs) refer to a new type of functional lipids obtained by chemically, enzymatically, or genetically modifying the composition and/or distribution of fatty acids in the glycerol backbone. Due to the unique physicochemical characteristics and health benefits of SLs (for example, calorie reduction, immune function improvement, and reduction in serum triacylglycerols), there is increasing interest in the research and application of novel SLs in the food industry. The chemical structures and molecular architectures of SLs define mainly their physicochemical properties and nutritional values, which are also affected by the processing conditions. In this regard, this holistic review provides coverage of the latest developments and applications of SLs in terms of synthesis strategies, physicochemical properties, health aspects, and potential food applications. Enzymatic synthesis of SLs particularly with immobilized lipases is presented with a short introduction to the genetic engineering approach. Some physical features such as solid fat content, crystallization and melting behavior, rheology and interfacial properties, as well as oxidative stability are discussed as influenced by chemical structures and processing conditions. Health-related considerations of SLs including their metabolic characteristics, biopolymer-based lipid digestion modulation, and oleogelation of liquid oils are also explored. Finally, potential food applications of SLs are shortly introduced. Major challenges and future trends in the industrial production of SLs, physicochemical properties, and digestion behavior of SLs in complex food systems, as well as further exploration of SL-based oleogels and their food application are also discussed.

KEYWORDS

structured lipids, synthesis, properties, health benefits, applications

1 | INTRODUCTION

Fats and oils are consumed in daily diets as an important source of energy, essential fatty acids, and fat-soluble nutrients. In food processing, they are widely used as a heat transfer medium and a major food ingredient rendering desirable texture, flavor, and mouthfeel to various lipid-based food products, such as margarine, mayonnaise, chocolate, infant formula, frozen dessert, and bakery products. Natural lipids are mainly composed of triacylglycerols (TAGs, ~98%) with several minor components, including monoacylglycerols (MAGs), diacylglycerols (DAGs), phospholipids, glycolipids, phytosterols, and free fatty acids (FFAs) (Shahidi, 2005). However, most lipids of natural origin have limited applications in their original state due to specific fatty acid and triacylglycerol composition (Ribeiro, Grimaldi, Gioielli,

& Gonçalves, 2009b). Although human milk fat and cocoa butter are among the few exceptions with inherently excellent functionality, neither of them can meet the substantial demand in the food industry for infant nutrition or chocolate products because of certain social and/or environmental factors. For example, infant nutrition from breastfeeding can not be fully satisfied due to mother's inability or option (Faustino et al., 2016) and likewise, natural cocoa butter may not be sufficient for chocolate products because of restricted climate, limited supply, and fluctuating price (Bahari & Akoh, 2018a). Furthermore, despite the great success of hydrogenated vegetable oils over the past 100 years, there are increasing health concerns and/or regulatory restrictions on the dietary ingestion of saturated fatty acids (SFAs) and *trans* fatty acids (TFAs) that have been found to be closely linked to cardiovascular diseases (Akoh, 2017; Shahidi, 2005).

Lipid modification has been proved to be a powerful tool for addressing the above challenges, where structured lipids with improved function and/or nutrition can be obtained by changing the fatty acid profiles (for example, chain length, unsaturation level, and positional distribution) of natural fats and oils (Kadhum & Shamma, 2017; Osborn & Akoh, 2002). In some earlier papers and book chapters, SLs were specifically defined as TAGs with modified fatty acid composition and/or positional distribution (Lee & Akoh, 1998; Osborn & Akoh, 2002; Shahidi, 2005); however, recently published works have discussed the definition of SLs in a broad sense where MAGs and DAGs are also included in the category of SLs (Akoh, 2017; Kim & Akoh, 2015). There are various SLs of commercial interest, such as medium- and long-chain TAGs (MLCTs), human milk fat substitutes (HMFSs), cocoa butter equivalents (CBEs), *trans*-free plastic fats, MAGs, and DAGs (Kim & Akoh, 2015), which have been summarized in Table 1. For example, MLCTs can be used to provide readily available energy by introducing medium-chain fatty acids (for example, caprylic acid (C8:0) and capric acid (C10:0); Abed et al., 2018; He, Li, Guo, & Chen, 2018). *Trans*-free plastic fats are synthesized mainly for replacing partially hydrogenated vegetable oils such as margarine and shortening fats (Lakum & Sonwai, 2018; Li et al., 2018b). HMFSs can be obtained with palmitic acids primarily at the *sn*-2 position and unsaturated FAs (for example, oleic acids) mainly at the *sn*-1,3 positions to mimic human milk fat, which is the "gold standard" (Şahin-Yeşilçubuk & Akoh, 2017; Zou, Pande, & Akoh, 2016a). The incorporation of long-chain polyunsaturated fatty acids (PUFAs, for example, *n*-3 series) is intended to improve the nutritional value of lipids thanks to the physiological function of these kinds of fatty acids (Ganesan, Brothersen, & Mcmahon, 2014; Patterson, Wall, Fitzgerald, Ross, & Stanton, 2012). Furthermore, synthesis of 1,3-DAGs has been an ongoing research interest mainly due to their potential health benefits in the reduction of serum TAG/cholesterol level and body fat accumulation as well as in the modulation of glucose metabolism (Lee et al., 2019; Lo, Tan, Long, Yusoff, & Lai, 2008; Phuah et al., 2015).

When a new type of SL is produced, it is essentially important to have a basic understanding of its physicochemical properties, such as solid fat content (SFC), crystallization and melting behavior, rheological property, interfacial property, and oxidative stability, which largely determine their food applications. It is widely accepted that the physical and chemical characteristics of SLs are mainly dependent on their chemical structure and also affected by processing conditions. Take margarine fat for example, it requires a particular SFC-temperature profile to ensure product functionality, namely, a 15 to 35% of SFC at room temperature for good spreadability, no less than 10% at 20 °C avoiding oil loss, and a 2 to 3% of SFC at 33.3 °C together with sharp melting near the body temperature to impart cooling sensation in the mouth. Moreover, small needle-like β'-form crystals with size not exceeding 5 μm are preferred to confer smooth texture to the final products and avoid graininess from larger crystal size (30 to 140 μm). All the above-mentioned requirements can be achieved by manipulating chemical composition (for example, a diverse composition of FAs and TAGs) and processing conditions (for example, a lower crystallization temperature, a rapid cooling rate, and applied shear; Osborn & Akoh, 2002). In addition to physical properties, the oxidative stability of SLs also needs to be assessed as a function of chemical structure and processing conditions (Shahidi & Zhong, 2010). Compared to unmodified fats and oils, SLs are produced by applying extra processing and purification steps with altered fatty acid profiles and even acylglycerol composition (Osborn & Akoh, 2002; Sproston & Akoh, 2016b; Yeoh et al., 2014). Furthermore, when they are applied in the form of emulsions (Diao, Guan, Zhao, Chen, & Kong, 2016; Kabri et al., 2013; Zou & Akoh, 2015b), SLs may exhibit a more complicated behavior regarding oxidative stability, which need to be fully understood and controlled for extended shelf-life and acceptable quality.

The health aspects of SLs are another major consideration when developing SLs and SL-based products. It is recognized that the nutritional values of SLs primarily result from their specific fatty acid composition (for example, PUFA-enriched SLs) and/or positional distribution as well as the resultant metabolic characteristics (for example, MLCTs, HMFSs, and 1,3-DAGs), whereas *trans*-free plastic fats mainly contribute to reduced ingestion of TFAs, thereby avoiding their adverse effects. In recent years, oleogels (also called physically structured oils), which possess solid-like property and retain nutritional profile of liquid oil (Agregan et al., 2019), have been considered to be a promising strategy to fully or partially replace saturated and/or *trans* fats and even interesterified fats of palm origin and have attracted considerable research interest (Demirkesen & Mert, 2019; Gravelle & Marangoni, 2018;

TABLE 1 Fatty acid composition, acylglycerol composition, and major characteristics of common SLs

SL Type	Fatty acid composition	Acylglycerol composition	Major characteristics	References
Trans-free plastic fats	• Diverse fatty acid composition • Increasing the saturated fatty acid content by interesterification between liquid oil and palm-based oils • Zero/low *trans* fatty acids	Diverse TAG species	• 15% to 35% SFC at room temperature for good spreadability, and no less than 10% at 20 °C avoiding oil loss • Small fine needle-like β' crystals confer smooth texture to the products. • Desirable crystal size no more than 5 μm (larger size (30 to 140 μm) may lead to graininess).	Norizzah, Chong, Cheow, and Zaliha (2004); Pande and Akoh (2013); Rao, Sankar, Sambaiah, and Lokesh (2001); Ribeiro et al. (2009a)
CBEs	• Oleic acids at the *sn*-2 position • Saturated fatty acids (for example, palmitic acids and stearic acids) at the *sn*-1,3 positions	1,3-Dipalmitoyl-2-oleoylglycerol (POP, 13.6% to 15.5%) 1-Palmitoyl-2-oleoyl-3-stearoylglycerol (POS, 33.7% to 40.5%) 1,3-Distearoyl-2-oleoylglycerol (SOS, 23.8% to 31.2%)	• Solid like at room temperature (SFC > 50%), and melt rapidly in the mouth. • Desirable polymorph is β form.	Biswas, Cheow, Tan, and Siow (2018); Yamoneka et al. (2018b)
HMFSs	• Palmitic acids mainly at the *sn*-2 position • Unsaturated fatty acids (for example, oleic acids) at the *sn*-1,3 positions	1,3-Dioleoyl-2-palmitoylglycerol (OPO)	• Promoting the adsorption of palmitic acids and calcium.	Turan, SAhin YeşIlçUbuk, and Akoh (2012); Zou, Jin, Guo, Xu, and Wang (2016b)
MLM-type	• LPUFAs at the *sn*-2 position • Medium fatty acids (for example, caprylic acids and capric acids) at the *sn*-1,3 positions	MLM	• Medium fatty acids released by pancrelipase in the small intestine can be directly absorbed and provide readily energy in the liver. • 2-MAG produced can form chylomicrons that are adsorbed through lymphatic systems, which is beneficial for malabsorption patients. • MLM-type SLs contain fewer calories (21 to 20 kJ/g) compared to conventional fats and oils (38 kJ/g).	Bandarra et al. (2016); Nunes, Pires-Cabral, and Ferreira-Dias (2011); Speranza and Macedo (2012); Wu, Zaniolo, Schuster, Schlotzer, and Pradelli (2017)
MAGs	—	1 (3) -MAG, and 2-MAG	• Nonionic surfactants capable of using as emulsifiers in the food industry	Feltes et al. (2013); Norn (2015)
DAGs	—	1,2-DAG, and 1,3-DAG	• Nonionic surfactants capable of using as emulsifiers in the food industry • 1,3-DAGs have been reported to reduce serum TAG level and suppress body fat accumulation.	Feltes et al. (2013); Lo et al. (2008); Matsuo (2007); Norn (2015)

Martins, Vicente, Cunha, & Cerqueira, 2018; Patel, 2017a; Scholten, 2019; Singh, Auzanneau, & Rogers, 2017). Most recently, Akoh's group has developed novel oleogels using SLs as the solvent, which combines the health benefits of both SLs and oleogels and poses a unique direction for exploring novel food applications of SLs in the near future (Willett & Akoh, 2019a, 2019b). Furthermore, recent interests have also been focused on delaying lipid digestion through a food structuring strategy to curb lipid-related health issues (for example, obesity) (Corstens et al., 2017; Guo, Ye, Bellissimo, Singh, & Rousseau, 2017; Sarkar, Zhang, Holmes, & Ettelaie, 2019).

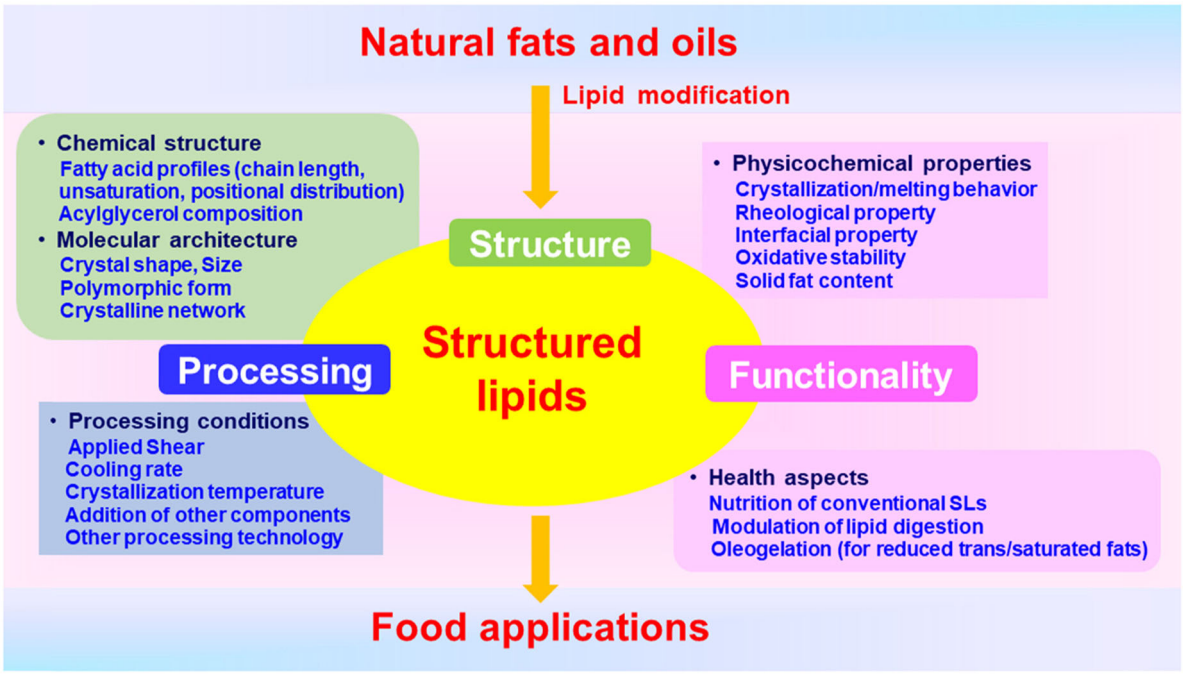

FIGURE 1 Schematic representation of the main content of the present review

Although there have been several reviews either on a particular type of SLs such as HMFSs (Şahin-Yeşilçubuk & Akoh, 2017; Wei, Jin, & Wang, 2019; Zou et al., 2016a), MAGs and/or DAGs (Feltes, De Oliveira, Block, & Ninow, 2013; Lee et al., 2019; Lo et al., 2008; Phuah et al., 2015), MLM-type SLs (Utama, Sitanggang, Adawiyah, & Hariyadi, 2019), and *trans*-free plastic fats (Berger & Idris, 2005; Pande & Akoh, 2013), or on an overview of several common SLs (Ferreira & Tonetto, 2017a; Kim & Akoh, 2015; Rohm, Schäper, & Zahn, 2018), very few of them have focused on processing-structure-function relationship of the corresponding SLs. An early review of Osborn and Akoh (2002) gave a concise introduction to several important physical and nutritional properties of SLs and their related applications. Recently, Rohm et al. (2018) have summarized the application of interesterified fats (that is, CBEs and *trans*-free plastic fats) in chocolate and bakery products and shortly introduced a few physical properties including SFC and crystallization behavior. On the basis of the latest advances in SLs and several novel lipid-based products (for example, oil foams, emulsions, and oleogels), the current work provides a comprehensive review of SLs in terms of their synthesis methods, physicochemical properties, and health aspects (Figure 1). First, novel lipase immobilization techniques and their applications for producing SLs were discussed, with a short introduction to the genetic engineering approach. Some important physicochemical properties of SLs were also presented, such as SFC, crystallization and melting behavior, rheological property and interfacial property, as well as their oxidative stability as influenced by chemical structures and processing conditions. In regard to the heath aspects of SLs, our emphasis was mainly put on modulation of lipid digestion by biopolymers and oleogelation of liquid oils for reduced *trans*/saturated fats, while nutritional values of conventional SLs due to their unique fatty acid profiles and metabolic characteristics were also briefly summarized. Finally, potential applications of SLs in the food industry were shortly introduced.

2 | SYNTHESIS OF SLS

Various approaches including chemical, enzymatic, and genetic methods can be applied to produce SLs, among which chemical interesterification is a traditional and common approach in practical production due to its low cost, ease of handling, and scaling up (Akoh, 2017; Osborn & Akoh, 2002). However, the disadvantages of the chemical method cannot be neglected, such as lack of specificity and selectivity, unwanted side products, higher energy consumption as well as thermal damage of the reactants and products (Shahidi, 2005). In this regard, both the scientific and industrial communities have endeavored to find an alternative method (that is, enzymatic approach). Comprehensive knowledge of enzymes in lipid modification can be observed in a recent review by Bornscheuer (2018). Lipases have been the most widely used enzymes in the production of SLs with considerable advantages such as chemo-, regio-, and stereo-selectivity, mild reaction conditions, and environmentally friendly (Bornscheuer, 2018). They can be categorized as *sn*-1,3-specific and non-specific enzymes and are capable of catalyzing a variety of

TABLE 2 Commonly used enzymatic approaches for production of SLs

Synthetic methods	Description	Reaction diagram
Direct esterification	Involving esterification between FFAs and glycerols	R_1-CO-OH+R-OH → R_1-CO-OR +H_2O
Interesterification	Involving intra- and inter-molecular exchange of acyl groups of acylglycerol molecules	R_1-CO-OR_2+R_3-CO-OR_4 → R_1-CO-OR_4 + R_3-CO-OR_2
Acidolysis	Involving ester-exchange between TAGs and FFAs	R_1-CO-OR+R_2-CO-OH → R_2-CO-OR + R_1-CO-OH
Glycerolysis	Involving transesterification reaction between TAGs and glycerols to produce MAGs and DAGs	R-CO-OR_1+R_2-OH → R-CO-OR_2 + R_1-OH

reactions including direct esterification, interesterification, acidolysis, and glycerolysis (Table 2). Important technical and economic considerations when applying these types of reactions to produce various SLs can be found in numerous book chapters and review papers (Feltes et al., 2013; Ferreira & Tonetto, 2017a; Phuah et al., 2015; Soumanou, Perignon, & Villeneuve, 2013; Utama et al., 2019). Depending on product requirement and substrate type, various kinds of SLs including *trans*-free plastic fats, CBEs, HMFSs, MLCTs, MAGs, and DAGs can be produced by adopting suitable methods. A recent review of Kim and Akoh (2015) has summarized the enzymatic synthesis of these types of SLs based on published studies between 2010 and 2014. Therefore, we simply give an updated progress on lipase-catalyzed synthesis of different SLs (Table 3).

In light of the substantial potential of immobilized enzymes for scale-up production of SLs, we primarily focus on the most recent progress on lipase immobilization techniques and their application for producing various SLs. In addition, a genetic engineering approach is also shortly introduced.

2.1 | Lipase immobilization technique

Many commercial lipases for the production of SLs are used in their immobilized forms (for example, Lipozyme TL IM, Lipozyme RM IM, Lipozyme 435, and Novozym 435), which makes it possible to produce SLs on an industrial scale. Compared to free lipases, their immobilized counterparts can be obtained through physical adsorption, physical entrapment, covalent bonding, and chemical crosslinking, with improved stability against heat and pH, easy recovery and reusability, and suitability for continuous processes (Facin, Melchiors, ValéRio, Oliveira, & Oliveira, 2019; Mateo, Palomo, Fernandez-Lorente, Guisan, & Fernandez-Lafuente, 2007). For physical immobilization, lipases can be immobilized onto the supports through weak interactions including van der Waals forces, hydrogen bonds, and hydrophobic interactions, or can be confined inside the support matrix, whereas chemical immobilization generally involves the formation of covalent bonds between the amino groups of lipases and the functional groups of the carriers (Facin et al., 2019; Shuai, Das, Naghdi, Brar, & Verma, 2017).

Considering their substantial advantages (for example, enhanced stability and reusability) over free lipases, immobilized lipases have been preferred by the food industry to produce various types of commercial SL products such as CBEs, *trans*-free margarine and shortening fats, and HMFSs for infant formulas (Dicosimo, Mcauliffe, Poulose, & Bohlmann, 2013). It has been estimated that the production scale of interesterified fats and oils, for instance, can reach 10^5 tons per year (Dicosimo et al., 2013). Despite the available commercial immobilized enzymes, continuous researches have been dedicated to designing novel carriers to immobilize lipases, intending to improve their performance further. For example, a recent work of our group has developed polysaccharide-functionalized magnetic microspheres (Fe_3O_4@SiO_2@{chitosan (CHI)/hyaluronan (HA)}$_3$) to immobilize lipase for the synthesis of 1,3-dioleoyl-2-palmitoylglycerol (OPO) (Cai et al., 2019). The lipase was covalently bonded onto the carrier through the EDC/NHS reaction, which catalyzes the coupling between carboxyl group of HA and amino group of lipases (Figure 2a). This immobilized lipase generally showed better thermal and storage stability than the free lipase and the commercial immobilized enzyme (Lipozyme RM IM), and it remained 85% of its initial activity after nine consecutive cycles. Various other carriers have also been fabricated to physically or chemically immobilize lipases for production of different SLs, which is summarized in Table 4.

When applying immobilized enzymes for the industrial production of SLs, several factors have to be considered to fulfill their catalytic potential and maximize the acquisition of the target products. First, for a specific SL, it is required to screen the desired immobilized lipase and determine the optimal processing conditions (for example, substrate ratio, enzyme amount, reaction temperature, time, and pH) (Bassan et al., 2019b; He et al., 2017c; Wang, Wang, Wang, Jin, & Wang, 2018b). Second, it is also important to select suitable reactors. Compared to stirring batch reactors which are mostly used in scientific researches, continuous fixed bed reactors are more common in an industrial setting for large-scale production of SLs with a superior efficacy (De Paula, Nunes, De Castro, & Dos Santos, 2018; Kim & Akoh, 2006; Wang et al., 2016). Xiao et al. (2018) have prepared immobilized

TABLE 3 An updated summary of enzyme-catalyzed synthesis of major SLs of commercial interest

SLs type	Synthesis methods	Substrates	Enzymes used	References
Trans-free plastic fats	Interesterification	Soybean oil + palm stearin + coconut stearin	Lipozyme RM IM (*Rhizomucor miehei*)	Lakum and Sonwai (2018)
		Soybean oil + fully hydrogenated palm oil	Lipozyme 435	Li et al. (2018b)
		Coix seed oil + fully hydrogenated palm oil + *Cinnamomum camphora* seed oil	Lipozyme RM IM (*Rhizomucor miehei*)	Xu et al. (2018)
		Acer truncatum oil + palm stearin + palm kernel oil	Lipozyme RM IM (*Rhizomucor miehei*)	Hu et al. (2017)
CBEs	Interesterification	*Irvingia gabonensis* seed fat + rapeseed oil/groundnut oil/palm super olein/*Dacryodes edulis* pulp oil	Lipozyme TL IM (*Thermomyces lanuginosa*)	Yamoneka et al. (2018b)
		Illipe butter + palm midfraction	Lipozyme RM IM (*Rhizomucor miehei*)	Bahari and Akoh et al. (2018a, 2018b)
		Palm mid-fraction + palm kernel oil + palm stearin + stearin acid + oleic acid	Lipozyme TL IM (*Thermomyces lanuginosa*)	Biswas et al. (2018)
HMFSs	Acidolysis	Fungal oil (from *Mortierella alpina* ALK, containing 47.7% arachidonic acid) + fractionated palm stearin (rich in tripalmitin) + oleic acid	Lipozyme RM IM (*Rhizomucor miehei*)	Wang et al. (2019)
		Microalgae oil (from *N. oculate,* rich in palmitic acids at the sn-2 position) + *n*-3 PUFAs (from *I. galbana*)	Novozym 435 (*Candida antarctica*)/Lipozyme 435 (*Candida antarctica*)/Lipozyme TL IM (*Thermomyces lanuginosus*)/Lipozyme RM IM (*Rhizomucor miehei*)	He et al. (2017b)
		Tripalmitin + FFAs (from camelina oil, rich in linolenic acid)	Heterologous *Rhizopus oryzae* lipase (rROL)/Lipozyme RM IM (*Rhizomucor miehei*)	Faustino et al. (2016)
	Interesterification	Tripalmitin + ethyl oleate	Lipase (*Candida parapsilosis*, immobilized on Accurel MP 1000)	Tecelão, Perrier, Dubreucq, and Ferreira-Dias (2019)
		Fractionated palm stearin + fish oil	Lipozyme TL IM (*Thermomyces lanuginosus*)	Ghosh, Sengupta, Bhattacharyya, and Ghosh (2016)
		Tripalmitin + extra virgin olive oil + flaxseed oil	Lipozyme TL IM (*Thermomyces lanuginosus*)	Ilyasoglu (2013)
		Tripalmitin + stearidonic acid-enriched soybean oil	Lipozyme TL IM (*Thermomyces lanuginosus*)/Novozym 435 (*Candida antarctica*)	Teichert and Akoh (2011)
MLM-type	Acidolysis	Grapeseed oil + caprylic acid (C8:0) or capric acid (C10:0)	Lipozyme TL IM (*Thermomyces lanuginosus*)/Lipozyme RM IM (*Rhizomucor miehei*)/Novozym 435 (*Candida antarctica*)	Bassan et al. (2019a)
		Microbial oils (from *Mortierella alpine,* rich in arachidonic acid) + caprylic acid (C8:0)	Lipozyme RM IM (*Rhizomucor miehei*)/Lipozyme TL IM (*Thermomyces lanuginose*)/Novozym 435 (*Candida antarctica*)/Lipozyme 435 (*Candida antarctica*)	Abed et al. (2018)

(Continues)

TABLE 3 (Continued)

SLs type	Synthesis methods	Substrates	Enzymes used	References
		Mustard+capric acid (C10:0) + caprylic acid (C8:0) + lauric acid (C12:0)	Lipozyme RM IM (*Rhizomucor miehei*)/Lipozyme TL IM (*Thermomyces lanuginosus*)/Novozym 435 (*Candida antarctica*)	Sengupta, Roy, Mukherjee, and Ghosh (2015)
	Ethanolysis + esterification	2-MAGs (obtained by ethanolysis of microalgae oil from *Isochrysis galbana*) + caprylic acid (C8:0)	Lipozyme TL IM (*Thermomyces lanuginosus*)	He et al. (2018)
		2-MAGs (obtained by ethanolysis of cod liver oil or tuna oil) + caprylic acid (C8:0)	Novozym 435 (*Candida antarctica*)/Lipozyme RM IM (*Rhizomucor miehei*)/Lipases D and DF (*Rhizopus oryzae*, immobilized on Accurel MP1000)/Lipase QLM without immobilization	Del Mar Muñío, Robles, Esteban, González, and Molina (2009)
MAGs	Glycerolysis	Anchovy oil + glycerol	Lipase PS-DI (*Burkholderia cepacia*)	Palacios et al. (2019)
		Sardine oil + glycerol	Lipozyme 435 (*Candida antarctic*)	Solaesa, Sanz, Falkeborg, Beltrán, and Guo (2016)
	Ethanolysis	Tuna oil + ethanol	Novozym 435 (*Candida antarctica*)/Lipozyme 435 (*Candida antarctica*)/Lipozyme RM IM (*Rhizomucor miehei*)/Lipozyme TL IM (*Thermomyces lanuginosus*)	Zhang et al. (2018a)
		Algal oil (*Schizochytrium sp.*) + ethanol	Novozym 435 (*Candida antarctica*)/Lipozyme 435 (*Candida antarctica*)/Lipozyme RM IM (*Rhizomucor miehei*)/Lipozyme TL IM (*Thermomyces lanuginosus*)/NS 40086 (*Aspergillus oryzae*)	Zhang et al. (2018b)
DAGs	Glycerolysis	Algae oil (from *Schizochytrium sp.*) + glycerol	Lipozyme 435 (*Candida antarctica*)/Lipozyme TL IM (*Thermomyces lanuginosus*)/Lipozyme RM IM (*Rhizomucor miehei*)	Wang et al. (2018a)
		Palm stearin/palm oil + glycerol	Lipozyme TL IM (*Thermomyces lanuginosus*)	Xu and Cao (2017b)
		Soybean oil + glycerol	Lipozyme 435 (*Candida antarctica*)	Zhang et al. (2016)
	Esterification	Oleic acid + glycerol	Lipase (*Rhizopus oryzaes*, immobilized on nanosized magnetite particles)	Zhao, Wang, Lin, Yang, and Wu (2019)
		α-Linolenic acid + glycerol	Lipase PCL (immobilized on ECR1030 and ECR8806 resins)	Liu et al. (2018)
		MAGs + caprylic acid (C8:0)	Novozym 435 (*Candida antarctica*)	Li et al. (2018a)
		FAs (a mixture of 60 wt% palm oil deodorized distillate and 40 wt% oleic acid) + glycerol	Lipozyme 435 (*Candida antarctica*)	Liu et al. (2016)
	Hydrolysis	Soybean oil + water	A lipase (from *Rhizopus oryzae* by recombinant *Pichia pastoris*)	Li et al. (2015a)
		Palm stearin + water	T1 lipase (expressed in recombinant *Escherichia coli*)	Xu, Guo, Wang, Wang, and Yang (2013)
	Interesterification	Soybean oil + distilled saturated MAGs	Lipozyme TL IM (*Thermomyces lanuginosus*)	Chen et al. (2017b)

TABLE 4 Immobilized lipases by using novel carriers and their applications in the synthesis of SLs

SLs type	Substrates	Lipase source	Carriers	Immobilization techniques	Immobilization methods	Performance	Reference
HMFSs (OPO)	Tripalmitin with oleic acid	*Rhizomucor miehei* lipase (RML)	Fe_3O_4@SiO_2 @{CHI/HA}$_3$	Chemical bonding	Lipase was immobilized onto Fe_3O_4@SiO_2@{CHI/HA}$_3$ via formation of amide bonds between the amino group of lipases and the carboxyl group of HA on the support by using EDC/NHS chemistry.	• The immobilized lipase showed better thermal stability retaining 53.9% of its initial activity than free lipase (47.7%) and commercial Lipozyme RM IM (50.1%) after storage at 60 °C for 48 hr, and better storage stability with 78% of its initial activity compared to free lipase (45%) and lipozyme RM IM after being stored at 4 °C for 30 days. • The immobilized lipase could be easily separated by applying an external magnetic field, and retain 85% of its initial activity after nine cycles.	Cai et al. (2019)
LML-type	Tricaprylin with ethyl linoleate	*Talaromyces thermophilus* lipase (TTL)	AB-8 resin (selected from five resins products: AB-8, DA201, ECR1030, SA-1 and HP2MGL)	Physical adsorption	TTL lipase was immobilized onto the AB-8 resin through hydrophobic interactions.	• TAGs with two long- and one medium-chain FAs content as high as 52.86 mol% were obtained. • The immobilized lipases showed good industrial potential as proved by scale-up reactions.	Lian, Wang, Tan, Wang, and Wang (2019)
Phytosterol esters	*β*-sitosterol with FAs (that is, oleic acid, linoleic acid, linolenic acid, and conjugated linoleic acid), or TAGs (that is, rapeseed oil, sunflower oil, linseed oil, and soybean oil)	*Candida rugosa* lipase (lipase AYS)	Mesoporous carbon spheres (MCS)	Physical adsorption	Immobilized lipase (Lipase@MCS) was obtained by immobilizing lipase onto MCS through hydrophobic interactions and cation-exchange interactions.	• Lipase@MCS served as both the emulsifier and catalyst with excellent thermal stability (achieving a conversion of 94.9% at 65 °C) and recyclability (a conversion up to 96.5% at 55 °C after 10 cycles). • All reactions showed more than 90.2% conversion within 1 hr for esterification and 1.5 hr for trans-esterification.	Dong et al. (2019)

(Continues)

TABLE 4 (Continued)

SLs type	Substrates	Lipase source	Carriers	Immobilization techniques	Immobilization methods	Performance	Reference
DAGs	α-linolenic acid or ethyl linolenate with glycerol	*Penicillium Camembertii* lipase (PCL)	ECR8806 resin (selected from six resin products: AB-8, D380, DA201, XAD1180N, ECR1030 and ECR8806)	Physical adsorption	Lipase PCL was immobilized onto the ECR8806 resin through hydrophobic interactions.	• The immobilized PCL exhibited higher thermal stability than its free form. • An 89.24% of esterification degree could be achieved with a DAG content of 54.49% under optimal conditions. • The immobilized lipase retained 91.60% of its initial activity after 10 cycles.	Liu et al. (2018)
Phytosterol esters	β-sitosterol with FAs (that is, oleic acid, linoleic acid, and linolenic acid), or TAGs (that is, linseed oil, rapeseed oil, and sunflower oil)	*Candida rugosa* lipase (AYS)	Hierarchically porous cellulose acetate monolith (CA-MN)	Physical entrapment	A monolithic continuous flow bioreactor (MCFB) containing CA-MN was first constructed, and lipase was physically entrapped by the porous structure of CA-MN.	• A more than 90% conversion could be obtained by using MCFB. • The kinetic parameters V_m/K_m and the catalysis effect increased by 66.4-fold and 6.1-fold, respectively, compared with conventional batch reactions. • MCFB could work continuously for 200 hr without loss of catalytic activity.	Xiao et al. (2018)
DAGs	Soybean oil with glycerol	*Rhizomucor miehei* lipase (RML)	Organic functionalized SBA-15 silicas	Physical entrapment	—	• Organic functionalization decreased the pH sensitivity of immobilized RML (pH 4.0 to 8.0, at 40 °C), and improved their thermal stability. • 1-Isocyanatopropane modified support immobilized RML showed a higher TAG conversion (82.99%) and DAG content (59.03%) than free RML (21.28% and 15.45%), comparable to commercial immobilized RML (NS40086, 78.30% and 60.30%)	Zhong, Chen, Liu, and Chen (2019)

(Continues)

TABLE 4 (Continued)

SLs type	Substrates	Lipase source	Carriers	Immobilization techniques	Immobilization methods	Performance	Reference
1,3-DAGs	Oleic acid with glycerol	*Rhizopus oryzae* lipase (ROL)	Aldehyde group-modified nanosized magnetite particles (NSM)	Chemical bonding	Lipase ROL was immobilized on the aldehyde group-modified NSM through chemical bonding between the amino group of lipase and aldehyde group of support.	• The specific hydrolytic and esterification activities of the immobilized lipase were 1,660% and 260% of those of the free lipase, respectively. • The immobilized lipase retained ~70% residual activity after 55 cycles. • Under optimal conditions, the content of 1,3-DAG could reach >76% without purification.	Zhao et al. (2019)
Trans-free plastic fats	Rice bran oil with palm stearin	*Candida rugosa* lipase	Ionic liquid (IL)-functionalized core-shell magnetic silica composites	Physical adsorption	Lipase was physically adsorbed onto the support by electrostatic interaction, hydrophobic interaction and hydrogen bond between IL groups and lipase.	• The immobilized lipase could be easily separated by an external magnetic field. • The interesterification degree reached 74.8% after four cycles.	Xie and Zang (2018)
HMFSs (OPO)	Tripalmitin with oleic acid	*Candida lipolytic* lipase (CLL)	Magnetic multi-walled carbon nanotubes (mMWCNTs)	Physical adsorption	Lipase was immobilized onto the mMWCNTs through hydrophobic and cation-exchange interactions between carboxylic acid of the support and lipase.	• The immobilized lipase shows better thermal stability than its free form, and retained 66.8% of its initial activity after 20 cycles.	Zheng et al. (2017)
Trans-free SLs	Soybean oil with palm stearin	*Candida rugosa* lipase	Aminopropyl-functionalized hydroxyapatite-encapsulated-γ-Fe_2O_3 nanoparticles	Chemical crosslinking	The lipase with the amino group was crosslinked with the amino group-modified support by using glutaraldehyde as a coupling reagent	• Under optimal reaction conditions, the stearoyl incorporation was 48.6%. • After four cycles, the stearoyl incorporation still reached 43.4% showing excellent reusability.	Xie and Zang (2017)

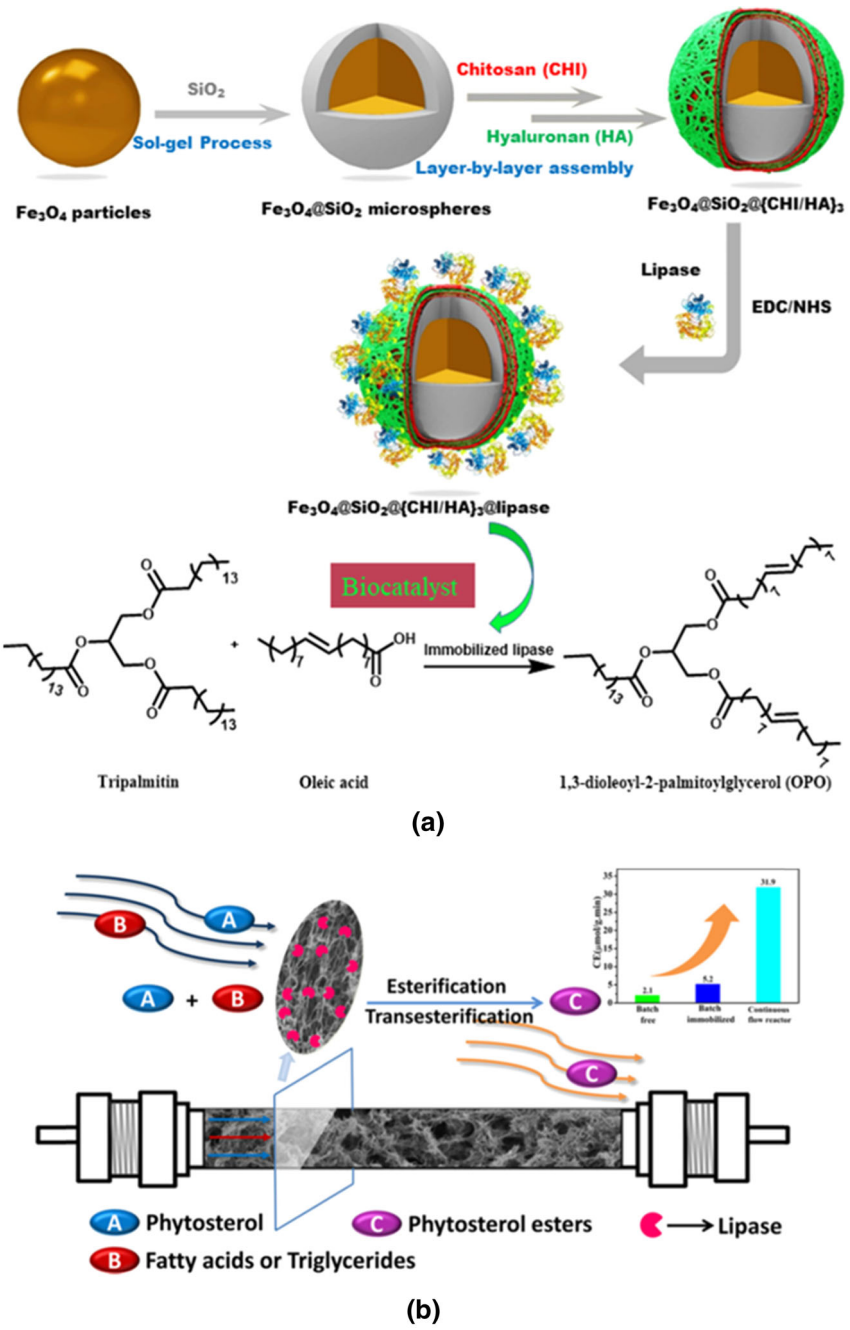

FIGURE 2 (a) Illustration of the construction of Fe_3O_4@SiO_2@{CHI/HA}$_3$-based on layer-by-layer self-assembly technique and its application for the immobilization of lipase and synthesis of OPO (Cai et al., 2019); (b) Illustration of a continuous flow bioreactor based on immobilized lipase using cellulose acetate monoliths as the support for production of phytosterol esters (Xiao et al., 2018)

lipase using a hierarchically porous cellulose monolith as the support, based on which a monolithic continuous flow bioreactor (MCFB) was constructed to produce phytosterol esters from phytosterol and oleic acid (Figure 2b). It has been found that the interesterification reaction in MCFB had the highest conversion at any molar ratio and temperature levels, followed by batch reactors with immobilized lipase and free lipase. The MCFB showed improved thermal stability, and under optimal conditions, the catalytic effect of MCFB reached 31.9 mmol/g·min, which was substantially higher than that of the immobilized batch reactor (5.2 mmol/g·min) and free lipase batch reactor (2.1 mmol/g·min). Furthermore, the MCFB enabled the reaction to be finished within 10 min and could work continuously for 200 hr without noticeable loss of catalytic activity (Xiao et al., 2018). In the commercial production of SLs by using fixed bed reactors, operation modes also play a significant role in affecting their production efficacy. For instance, when a single fixed bed reactor is used, the enzyme activity can be completely lost after operating long enough, and the step for conversion of new immobilized enzymes may reduce production efficiency. This problem can be overcome by adopting an operation mode where several fixed bed reactors are operated in series, and the exhausted reactor is recharged and then used as the last reactor in the series without disrupting the continuous production (Dicosimo et al., 2013).

In spite of the significant benefits of immobilized lipases as mentioned above, several limitations should not be ignored, such as possible enzyme activity loss upon immobilization, unfavorable variance in kinetic properties, mass transfer limitations as well as the cost of carrier and immobilization process (Dicosimo et al., 2013). It should also be noted that although immobilized lipases have been commercially utilized in the food industry (only by a few international food giants), the production cost is still relatively high compared to chemical methods. This has largely limited their food applications in a broader sense, especially when complex carriers or strategies are used for enzyme immobilization, which is, unfortunately, the common case in numerous published works. Furthermore, it is suggested that future researches should also focus on the real applicability to match a practical production setting when designing immobilized lipase. For example, despite the fact that magnetic-responsive immobilized lipase can be easily recovered, this advantage may be compromised in the presence of a continuous fixed bed reactor, which is commonly adopted for industrial production of SLs.

2.2 | Genetic engineering

The advancement of genetic engineering has made it possible to identify, isolate, modify, clone, and transfer desirable genes related to plant seed lipid production, achieving the synthesis of specific SLs in a more flexible manner (Zam, 2015). For example, biosynthesis of HMFSs in a model oilseed (*Arabidopsis thaliana*) has been achieved with more than 70% of palmitic acids at the *sn*-2 position by modifying the TAG metabolic pathway with genetic engineering technique (Van Erp et al., 2019). Vegetable oils with enhanced oxidative stability can be obtained from genetically modified cultivars by increasing oleic acid content and simultaneously reducing linolenic acid content (Wilkes, 2008). Most recently, high-oleic peanut oils have been available in the Chinese market that is gaining popularity. In North America, high-oleic canola oil has been largely used in commercial frying and food processing (Akoh, 2017). Genetic engineering can also be used to produce seed oils with a suitable amount of solid fat by lowering unsaturation, which can serve as plastic fats for the production of *trans*-free margarines and shortenings (Akoh, 2017; Osborn & Akoh, 2002). In addition to conventional oil crops, microorganisms including some fungi, algae, and bacteria have attracted research interest as an alternative source to marine fish for providing PUFAs, and this strategy can not only meet the increasing consumption demand for SLs containing PUFAs, but can also avoid potential food safety issue due to accumulation of heavy metals in fish oils (Béligon, Christophe, Fontanille, & Larroche, 2016; Gupta, Barrow, & Puri, 2012; Sijtsma & De Swaaf, 2004).

Several important concerns over genetic engineering techniques for practical production of SLs have to be taken into account, including high costs, long periods, and cross-pollination, and moreover the use of this method should not compromise the crop yield and hinder seed germination and growth. In addition, doubtful attitudes of consumers are still a major hurdle toward the edible use of genetically modified products. In contrast, the enzymatic approach has been a powerful and flexible tool to synthesize a large variety of SLs and furthermore, the scale-up production of SLs at reduced costs can be promising thanks to the development and application of immobilized lipase techniques.

3 | PHYSICOCHEMICAL PROPERTIES OF SLS

SLs are usually synthesized to achieve favorable physical properties for specific uses, which mainly depend on their chemical structure (that is, fatty acid chain, unsaturation level, and positional distribution) and molecular architecture (for example, polymorphic form, crystal shape, size, and the resultant crystalline network), and are also affected by processing conditions such as temperature, cooling rate, external force, and use of additives (Akoh, 2017; Osborn & Akoh, 2002). Furthermore, it is also important to understand their oxidative stability as affected by various factors including chemical structure, processing/purification steps, trace metals, and antioxidants for acceptable safety and quality of SL-based food products (Shahidi & Zhong, 2010).

3.1 | Solid fat content

Solid fat content (SFC) refers to the ratio (0 to 100%) of solid fat present in the oil at a specific temperature, which is usually determined by pulsed nuclear magnetic resonance (Shahidi, 2005). SFC of a fat system is mainly affected by fatty acid profiles, acylglycerol composition, and temperature. In general, the longer the chain length and the higher saturation degree the fatty acids, the larger SFC the fat (Shahidi, 2005). *Trans*-free plastic fats with increased SFC have been extensively produced through interesterification of liquid oil with solid fat (for example, palm stearin and fully hydrogenated oil) (Dian et al., 2017; Makeri, Sahri, Ghazali, Ahmad, & Muhammad, 2019; Zhu, Weng, Zhang, Wu, & Li, 2018a). Owing to the presence of hydroxyl (-OH) on the glycerol backbone, MAGs and DAGs show different acylglycerol composition compared to TAGs, thus affecting their SFC profiles. For example, according to SFC-temperature (*T*, 0–40 °C) curves, lard-MAG has been reported to exhibit the highest SFC over the temperature range, followed by lard-DAG and lard TAG (Cheong, Zhang, Xu, & Xu, 2009). Moreover, the SFC of the three samples all decreased with

TABLE 5 Possible crystal nucleation and growth modes inferred from Avrami exponent (Cheong et al., 2009)

Avrami exponent (n)	Possible mechanism of crystal nucleation and growth
3 + 1 = 4	Spherulitic growth from sporadic nuclei
3 + 0 = 3	Spherulitic growth from instantaneous nuclei
2 + 1 = 3	Disk-like growth from sporadic nuclei
2 + 0 = 2	Disk-like growth from instantaneous nuclei
1 + 1 = 2	Rod-like growth from sporadic nuclei
1 + 0 = 1	Rod-like growth from instantaneous nuclei

increasing temperature. Xu, Wei, Zhao, Chen, and Cao (2016) used palm oil (PO) and its derivatives (palm stearin, PS; palm mid fraction, PMF; palm olein, POL) to produce the corresponding DAG oils, and compared their SFC-*T* (0 to 50 °C) curves. The results showed that PS-DAG had a lower SFC than PS, whereas the other three kinds of DAG oils had a larger SFC at higher temperatures (10 to 50 °C), indicating that the SFC profiles may also be affected by oil types.

The change of SFC of an SL under supercooling conditions as a function of time (SFC–*t* curve) has been widely applied to monitor the isothermal crystallization kinetics. The mathematical relationship between SFC and time can be fitted by Avrami model (Litwinenko, Rojas, Gerschenson, & Marangoni, 2002):

$$\frac{\mathrm{SFC}\ (t)}{\mathrm{SFC}_{\max}} = 1 - \mathrm{e}^{-kt^n} \tag{1}$$

where SFC(t) and $\mathrm{SFC}_{\max}$ refer to the solid fat content at time t and the maximum SFC at the equilibrium, respectively. k is the Avrami constant, that is, the crystal growth rate constant, and n is the Avrami exponent defining the crystal growth mechanism. It should be noted that the Avrami exponent (n) is a parameter that is related to the time dependence of nucleation and the dimension number of crystal growth (Wright, Hartel, Narine, & Marangoni, 2000). For example, nucleation can be either instantaneous (where nuclei appears all at once at an early stage of the process) or sporadic (where the nuclei amount increases linearly with time), while crystal growth can take place as either rod-like, disc-like, or spherulitic type in one, two, or three dimensions (Wright et al., 2000; Zhu, Zhang, Wu, & Li, 2019). Possible crystal nucleation and growth modes inferred from the Avrami exponent have been summarized in Table 5 (Cheong et al., 2009). Another important parameter, half-time of crystallization ($t_{1/2}$), which indicates the time required to attain 50% of crystals, can be calculated from k and n (Metin & Hartel, 1998):

$$t_{1/2} = \left(\frac{\ln 2}{k}\right)^{1/n} \tag{2}$$

In a recent work of Zhu et al. (2019), fast-frozen special fat (IBSF) was produced from an interesterified blend of palm stearin and rapeseed oil. Based on Avrami analysis, they found that IBSF had lower k and higher $t_{1/2}$ values than the physical blend (PBSF), indicating a lower supercooling degree and slower crystallization rate of IBSF at the same temperature. Xu et al. (2016) reported that DAG-rich oils of palm origin crystallized faster than their corresponding TAG oils at the applied isothermal crystallization temperature (10, 15, 20, and 25 °C), with higher k and lower $t_{1/2}$ for DAG oils. In addition, all the palm-based DAG oils exhibited no spherulitic crystal growth with n less than 2, and compact structures of tiny needle-like crystals were observed from the microscopic images (Xu et al., 2016).

It should be pointed out that in some cases instead of SFC, another parameter crystallinity (%), which was calculated from crystallization heat by using DSC, was adopted to represent the amount of the formed crystals (Li et al., 2018b). Furthermore, the Avrami exponent (n) is not believed to be sufficient to make a definite conclusion about crystal growth mechanism unless combined consideration of the microscopical results (Saberi, Lai, & Toro-Vázquez, 2011).

3.2 | Crystallization and melting behavior

When fats and oils are cooled at a specific temperature, high-melting lipid molecules can first crystallize followed by those with lower melting points, leading to the formation of a multiscale crystalline structure (Acevedo & Marangoni, 2015; Marangoni et al., 2012). The crystallization process of lipids overall involves four phases including the occurrence of a thermodynamic driving force, nucleation, crystal growth, and recrystallization (Hartel, 2013). Once the crystalline structure is formed, it is equally important to know its melting property, which, in combination with crystallization behavior, greatly influences the physical properties of lipid-based food products. Various techniques such as differential scanning calorimetry (DSC), X-ray diffraction (XRD) and polarized microscopy (PLM) can be applied to characterize their crystallization and melting behaviors.

Fats and oils tend to crystallize in three major polymorphic forms, namely, α, β', and β, and their different characteristics have been presented in Table 6 (Wagh & Martini, 2017). For CBEs, the desired β crystals can impart favorable properties to chocolate products such as gloss, snap, flavor release, and mouthfeel. Several fats and oils including palm kernel oil, mango seed kernel fat, kokum butter, sal fat, shea butter, and illipe fat can be blended or modified to obtain cocoa butter equivalents, since they have similar fatty acid and triacylglycerol composition to cocoa butter and tend to yield β polymorph in a controlled process (Jahurul et al., 2013). For example, Bahari and Akoh (2018a) produced CBEs by enzymatically catalyzing a blend of illipe butter and palm midfraction at a ratio of 10:3 (w/w). The obtained interesterified products showed similar TAG profiles to the

TABLE 6 Major characteristics of polymorphic forms in SLs (Wagh & Martini, 2017)

Polymorphic form	Unit cell	Stability	Density	Melting point	X-ray diffraction spacing (Å)
α	Hexagonal	Least stable	Lowest	Lowest	4.15
β'	Orthorhombic	Metastable	Intermediate	Intermediate	3.8 and 4.2
β	Triclinic	Most stable	Highest	Highest	4.6

commercial cocoa butter (CB), with an onset melting temperature of 32.7 °C, predominant β polymorphs, and spherulitic crystals, which are comparable to those of CB. In regard to margarine and shortening fats, the formation of β' polymorph is preferred to confer smooth texture to the final products. It is widely accepted that a high diversity of fatty acid and triacylglycerol composition facilitates the formation of β' crystals (Pande & Akoh, 2013; Ribeiro, Basso, Grimaldi, Gioielli, & Gonçalves, 2009a). Interesterification generally led to an increase in TAG species. For example, in the work of Xu et al. (2018), new TAG peaks were observed after enzymatic interesterification of coix seed oil, fully hydrogenated palm oil and *Cinnamomum camphora* seed oil, compared to their raw material oils and physical blends. A similar result was also found by Li et al. (2018b), who reported new endothermic peaks in the DSC curves of interesterified oil (IO) from soybean oil and fully hydrogenated palm oil. IO exhibited similar melting curves to beef tallow (BT), composed of denser aggregated plate-like crystals mainly in the β' form with smaller sizes (10 to 25 μm).

MAGs have two isomers: 1-MAGs and 2-MAGs, the equilibrium ratio of which is 95:5 at room temperature (Krog & Sparsø, 2003). Vereecken et al. (2009) investigated the crystallization and melting behavior of saturated and unsaturated MAGs with different chain length as well as their two isomers by using DSC and XRD. For purified saturated MAGs (MAG ≥ 99%, consisting of 1-MAGs and 2-MAGs), three polymorphs including β, α, and sub-α with decreasing melting points were observed. Moreover, a second sub-α peak could be detected for the samples with longer chains (C18:0, C20:0, and C22:0; Vereecken et al., 2009). Similar results were also reported in a series of following studies of the same group (Verstringe, Danthine, Blecker, & Dewettinck, 2014; Verstringe, Danthine, Blecker, Depypere, & Dewettinck, 2013). As the chain length increased, the saturated MAG samples showed increasing crystallization and melting temperatures (Vereecken et al., 2009). For a mixture of monopalmitin and monostearin, however, the crystallization of α polymorph and the transition to sub-α form both occurred at a lower temperature compared to its pure constituents. In addition, only one sub-α polymorph was observed despite the presence of monostearin (C18:0), which was probably ascribed to the molecular incompatibility of the various MAGs (Verstringe et al., 2014). Krog (2001) also reported that MAG mixtures had lower crystallization and melting temperatures and a less complicated polymorphic behavior than their pure ones. Furthermore, taking 1-monostearin and 2-monostearin as an example, it has been found that 1-MAGs exhibited almost the same crystallization and melting behavior as the abovementioned purified samples, whereas 2-MAGs showed entirely different behavior, characterized by faster crystallization and only one β polymorph with greater stability (Vereecken et al., 2009), possibly due to the fact that the purified samples are mainly comprised of 1-MAG isomers (Krog & Sparsø, 2003). In contrast, unsaturated MAGs had a less complicated crystallization and melting behavior with only one peak in the crystallization and melting curves, and the XRD result demonstrated it to be the stable β polymorph (Vereecken et al., 2009). However, no definite conclusion could be reached for unsaturated MAGs, since previous work had observed more than one polymorphic form (Hagemann, 1988), which needs to be further studied. When the chain length was fixed, an increase in the unsaturation level (that is, monoolein, monolinolein, and monolinolenin) resulted in lower crystallization and melting temperatures (Vereecken et al., 2009). Recently, He et al. (2017a) produced fish oil-based MAGs containing different amounts of *n*-3 PUFAs, and they also reported that the crystallization temperature decreased with increasing *n*-3 PUFAs content in MAGs.

Similarly, there also exist two isomers for DAGs: 1,3-DAGs and 1,2-DAGs, with 1,3-DAGs being the dominant fractions (Lo et al., 2008). In general, 1,3-DAG molecules crystallize faster and the obtained crystals show a higher melting point than 1,2-DAGs with the same fatty acid composition, which is probably attributed to their different lamellar conformation where 1,3-DAGs and 1,2-DAGs form a V-shaped and a hairpin-shaped structure, respectively (Lo et al., 2008). As for DAGs with the same molecular conformation, the melting point increases with increasing fatty acid saturation, as observed in DSC results (heating rate: 1 °C/min) of purified 1,3-dioleoyl-*sn*-glycerol (26.33 °C), 1-palmitoyl-3-oleoyl-*sn*-glycerol (43.63 °C), and 1,3-dipalmitoyl-*sn*-glycerol (69.70 °C; Xu & Cao, 2017a). In the work of Saitou et al. (2012), for example, when DAG-rich oils (DAG: 85.0%) produced from rapeseed oils were cooled at 3 °C, the resultant crystals were mainly composed of 1,3-DAGs instead of 1,2-DAGs. In addition, the DAG fractions have been found to crystallize in a sequential manner with high-melting molecules such as 1,3-disaturated DAGs crystallizing first followed by 1,3-saturated-unsaturated and 1,3-diunsaturated

DAGs (Saitou et al., 2012). Furthermore, it has been reported that 1,3-DAGs tend to crystallize into β-form polymorph, while 1,2-DAGs usually exhibit α and β' form (Craven & Lencki, 2011; Saitou et al., 2012; Xu & Cao, 2017a). In palm-based DAG-rich oils (DAG: 87.43% to 92.61%) with 1,3-DAG/1,2-DAG ratio from 7/4.2 to 7/2.1, the XRD results showed that all the DAG oils predominantly consisted of β-form crystals (78.58% to 94.53%) as well as a small amount of β'-form polymorphs (5.47% to 21.42%) (Xu et al., 2016). Since 1,3-DAGs are the primary components of DAG fractions, the presence of DAGs in TAG oils or an increase in DAG content generally leads to an increase in the number of β crystals (Miklos, Zhang, Lametsch, & Xu, 2013; Saberi, Tan, & Lai, 2011; Zhao, Sun, Qin, Liu, & Kong, 2018).

Different processing parameters, including supercooling degree, cooling rate, external force, and use of additives, can be controlled to adjust the crystallization and melting behaviors of SLs. Generally, a high supercooling degree, fast cooling rate, and applied shear contribute to the formation of small fine crystals (Akoh, 2017; Shahidi, 2005). Crystallization temperature determines the degree of supercooling, which is one of the driving forces for crystallization (Metin & Hartel, 2005). With decreasing crystallization temperature (25 to 10 °C), palm-based TAG oils and their derived DAG oils all showed increased equilibrium SFC and higher crystallization rate (Xu et al., 2016). Similar observations were reported in interesterified fast-frozen special fats (Zhu et al., 2019). A slow cooling rate means that lipid molecules have more time to arrange themselves during crystallization and form more stable polymorphs (Metin & Hartel, 2005). In a hybrid system containing DAGs and TAGs, for example, the samples crystallizing at a slow cooling rate (2 °C/min) had a higher crystallization enthalpy and then transformed to a more stable form after subsequent isothermal crystallization, compared to those crystallizing at a rapid cooling rate (25 °C/min) (Tavernier, Norton, Rimaux, Lazidis, & Dewettinck, 2019b). Applying shear also has a significant effect on fat crystallization through various manners, such as accelerating fat nucleation, growth, and polymorphic transition, and inducing orientation of fat crystal networks (Tran & Rousseau, 2016). Furthermore, other processing techniques including high-intensity ultrasound (Kadamne, Ifeduba, Akoh, & Martini, 2017; Povey, 2017; Wagh, Birkin, & Martini, 2016) and high pressure processing (Zulkurnain, Maleky, & Balasubramaniam, 2016) have also been found to affect fat crystallization behavior and physical properties, but further research is still needed before these novel methods can be applied in an industrial setting. The crystallization behavior of SLs may also be altered by adding minor components. For instance, in DAG-rich oils (DAG > 80%) unpleasant "clouding" phenomenon usually occurs during storage as a result of crystallization of high-melting DAG fractions (Saitou, Homma, Kudo, Katsuragi, & Sato, 2014). This problem has been solved through the addition of polyglycerol fatty acid esters (PGFEs), mainly due to formation of a liquid-crystal-like supramolecular complex structure containing the high-melting DAG fractions and PGFEs (Saitou et al., 2014; Saitou et al., 2017). Other minor components such as phytosterol esters (Daels, Foubert, & Goderis, 2017), sucrose behenate (Domingues, Da Silva, Ribeiro, Chiu, & Gonçalves, 2016), lecithin (Rigolle et al., 2015), and sugar (West & Rousseau, 2017) have also been reported to adjust the crystallization and melting behavior of fats and oils. It should be noted when SLs (for example, MAGs and DAGs) are added as minor components into a TAG system, they show complex promotion or retardation effect on the crystallization and melting behavior, depending on additive type, addition amount, and bulk oil type (Alfutimie, Al-Janabi, Curtis, & Tiddy, 2016; Da Silva et al., 2017; Da Silva, Domingues, Chiu, & Goncalves, 2017b; Tavernier, Moens, Heyman, Danthine, & Dewettinck, 2019a), which has been extensively reviewed (Patel & Dewettinck, 2015b; Smith, Bhaggan, Talbot, & Van Malssen, 2011) and will not be discussed here.

3.3 | Rheological properties

Several SLs (for example, CBEs and *trans*-free plastic fats), which are solid like at room temperature, are mixtures where liquid oils are entrapped in a three-dimension network formed by solid fat crystals (Acevedo & Marangoni, 2015; Acevedo, Peyronel, & Marangoni, 2011). The mechanical properties of the fat crystalline network are defined by the amount of solid fat crystals, their size, shape, and structural organization, which further determine the texture of the fat-based products (Gonzalez-Gutierrez & Scanlon, 2018). Oscillatory shear rheology has been widely applied to characterize the mechanical properties of the fat crystalline network and to investigate the structure–function relationship of fat systems, which can be categorized as two modes: small amplitude and large amplitude oscillatory shear rheology (referred to SAOS and LAOS, respectively), according to the magnitude of the applied stress or strain (Gonzalez-Gutierrez & Scanlon, 2018; Macias-Rodriguez & Marangoni, 2018; Rigolle, Van Den Abeele, & Foubert, 2018). A typical oscillatory test measures an output of the stress (strain) wave in response to an input of a sinusoidal strain (stress) wave, and from the stress–strain curve various rheological parameters including complex modulus (G^*), phase angle (δ), storage modulus (G'), loss modulus (G''), and yield stress (σ_γ) can be obtained (Rao, 2010).

In a SAOS mode, the experiment is performed within the linear viscoelastic region (LVR), where the deformation is reversible and G' and G'' is independent on the applied strain or stress at a given temperature and frequency. For a fat system, a very small strain in the order 0.01% to 0.1% is usually applied, due to the weak Van der Waals forces that hold the fat crystal network (Macias-Rodriguez & Marangoni, 2018).

In this region, fats behave as a viscoelastic solid with high G' (~10^5 to 10^6 Pa) and low frequency dependence (Macias-Rodriguez & Marangoni, 2018). SAOS has been used to monitor the crystallization of SLs or SL-based products and to assess the mechanical strength of the formed crystalline network. Xu et al. (2016) reported that all palm-based DAG oils crystallized faster than their corresponding starting materials at 25 °C by plotting G^* as a function of time. Moreover, the equilibrium G^* of PMF-DAG and PO-DAG was higher than PMF and PO, respectively, suggesting the stronger network structure formed by these DAG oils. Saghafi, Naeli, Tabibiazar, and Zargaraan (2018) produced zero-*trans* cake shortening (ZTC) from an interesterified blend of palm stearin and canola oil, and compared its rheological properties with two commercial shortenings (CM101 and CM102). The frequency sweep (0.1 to 20 Hz) showed that the change in moduli for all the samples was independent of the frequency with $G' > G''$ indicating a solid-like crystalline network, but the G' value of ZTC was lower than that of CM101 and CM102. Furthermore, it can be seen from the temperature sweep test (5 to 60 °C) that the phase transition from solid-like to liquid-like behavior (where G' and G'' are equal and tan $\delta = 1$) occurred at a lower temperature for ZTC (47 °C) compared to CM101 (55 °C) and CM102 (48 °C), which was attributed to the lower SFAs and SFC in ZTC.

SAOS is also a powerful tool to elucidate the structure-function relationship of crystalline fat networks. Several rheo-physical models like the fractal model can be applied to quantitatively relate the linear viscoelastic properties of a fat system to its microstructure (Macias-Rodriguez & Marangoni, 2018; Tang & Marangoni, 2007). This model assumes that fat crystal network is composed of fractal flocs or aggregates formed by self-assembly of fat molecules, based on which the elastic modulus G' of the system can be related to its volume fraction Φ in a power-law manner:

$$G' \sim \Phi^{[(d+x)/(d-D)]} \quad \Phi < 0.1 \tag{3}$$

$$G' \sim \Phi^{[1/(d-D)]} \quad \Phi > 0.1 \tag{4}$$

where d is the Euclidean dimension of the embedding space (usually 3), x is the backbone fractal dimension (~1 to 1.3), and D is the fractal dimension, which is linked to spatial distribution, compactness, and morphology of the network, and is also used to explain the aggregation mechanism of the network. However, there are very few reports on its application in a fat system containing SLs, which need to be explored in future researches.

In contrast to SAOS, LAOS can provide nonlinear viscoelastic information of fat systems, which is of practical importance in the processing and application of fat-based products such as extrusion of shortening and spreading of butter (Macias-Rodriguez & Marangoni, 2018; Rodriguez, 2019). When the applied stress exceeds the yield stress, plastic flow occurs as a result of the breakdown of the weak interactions between fat crystals, and the fat network can restore due to the restructuring of the fat crystals once the stress is removed (Gonzalez-Gutierrez & Scanlon, 2018). Nevertheless, it should be noted that LAOS has not been extensively used in an SL system as SAOS, and the large deformation rheological behavior of SLs is traditionally studied by using the compression mode of a texture analyzer from which "hardness" or "firmness" can be obtained (Saghafi et al., 2018; Tavernier, Norton, et al., 2019b). Fortunately, the significance of LAOS in fully understanding the non-linear rheology of fat systems has aroused recent attention (Macias-Rodriguez & Marangoni, 2018), which needs to be further investigated to guide the practical application of new types of SLs.

3.4 | Interfacial properties

MAGs and DAGs are believed to be surface active due to the presence of hydroxyl groups (-OH) as well as fatty acid moieties in the molecules (Cheong et al., 2012). The addition of MAGs or DAGs into TAG oils has been found to be able to decrease the interfacial tension of the TAGs oil with MAGs being more effective (Shimada & Ohashi, 2003). Chen, Mcclements, and Decker (2014) reported that a small amount of 1,3-dioleins (0% to 2.5%, w/w) added to stripped soybean oil (SSO) only led to a slight decrease in interfacial tension, which was attributed to their lower interfacial tension instead of surface-active ability. The addition of monooleins at the same concentrations, however, significantly decreased the interfacial tension in a concentration-dependent manner, suggesting that MAGs were partly adsorbed at the SSO-water interface and the adsorbed amount increased with increasing concentrations until the maximum adsorption at about 1.5% (Figure 3a). Moreover, 7,7,8,8-tetracyano quinodimethane (TCNQ) spectrophotometry experiment confirmed that DAGs and MAGs added in the SSO did not form ordered structures at the given concentration range.

TAGs have been fully or partially substituted by DAGs to produced emulsions with enhanced nutrition (Diao et al., 2016; Long et al., 2015; Ng, Lai, Abas, Lim, & Tan, 2014). It has been demonstrated that variation in the oil phase significantly influences adsorption behavior of emulsifiers, viscoelastic properties of the formed interfacial film, and emulsion stability (Bergfreund, Bertsch, Kuster, & Fischer, 2018; Zare, Allison, & Mcgrath, 2016; Zhai et al., 2011). Long et al. (2015) constructed O/W emulsions stabilized by sodium caseinate with either peanut oil (PO) or peanut oil-based DAG oil (PO-DAG, 95.95% purity) as the oil phase. They observed that PO-DAG/water interface had a lower interfacial tension and almost remained constant, whereas the PO–water interface had a significantly higher initial interfacial tension and then decreased with the evolution

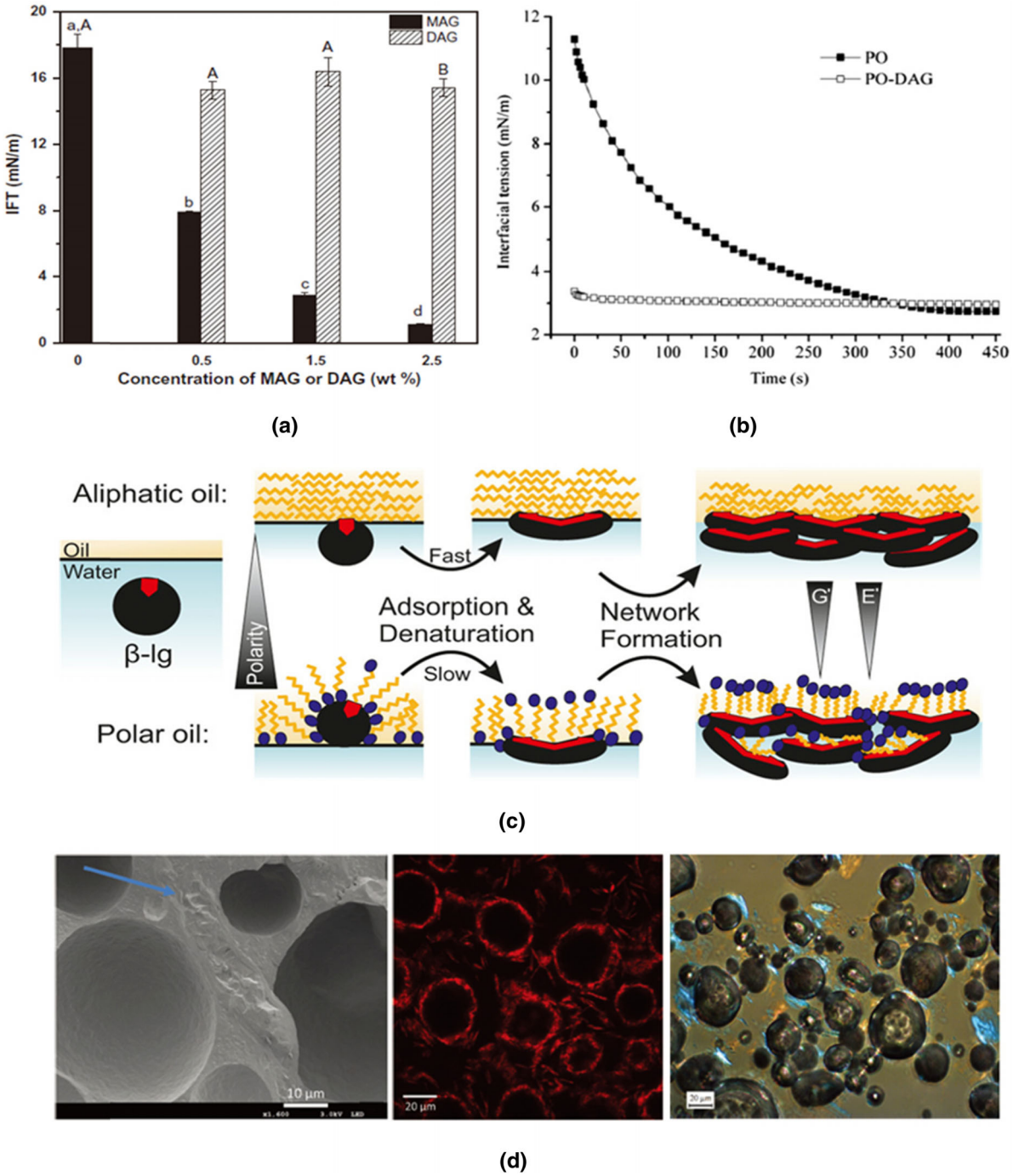

FIGURE 3 (a) The impact of addition of different concentrations of MAG or DAG on the interfacial tension of stripped soybean oil (SSO) (Chen et al., 2014); (b) The interfacial tension versus time for the 1 wt% sodium caseinate solution in the presence of peanut oil (PO) or peanut oil-based diacylglycerol (PO-DAG) (Long et al., 2015); (c) Schematic illustration of the effect of oil polarity on adsorption of β-lactoglobulin and interfacial film structure at oil–water interface in a model system (Bergfreund et al., 2018); (d) Microscopy images of oil foams formed by air bubbles dispersed in sunflower oil stabilized by MAGs: (left) cryo-scanning electron microscopy (SEM) image, (middle) CLSM image, and (right) polarized light image (Heymans et al., 2018)

of time. When protein adsorption equilibrium was reached, the interfacial tension of PO–water interface was slightly lower than that of the PO–DAG–water interface (Figure 3b). Similar results were also observed in a previous work by Sakuno, Matsumoto, Kawai, Taihei, and Matsumura (2008), where they compared the adsorption behaviors and structural changes of β–lactoglobulin at the DAG–water interface and TAG–water interface, and found that a larger concentration of proteins was adsorbed to the TAG–water interface with a higher degree of structure unfolding. In a recent work using a

model O/W emulsion, it has been reported that an increase in oil polarity delayed the adsorption and denaturation of proteins, thus leading to slower formation of interfacial film with decreased mechanical strength (Figure 3c; Bergfreund et al., 2018), which can largely explain the above-mentioned results when DAG oils are used as the oil phase in contrast to TAGs.

Oil foams, also called oleofoams, non-aqueous foams, or whipped oils, refer to a colloidal dispersion where air bubbles are dispersed in oils (Figure 3d; Heymans et al., 2018), which can be used as low-calorie food products (Gunes et al., 2017) and lubricating oil (Binks, Davies, Fletcher, & Sharp, 2010). Crystalline particles including MAGs (Gunes et al., 2017; Heymans et al., 2018), DAGs (Shrestha, Shrestha, Sharma, & Aramaki, 2008; Shrestha, Shrestha, Solans, Gonzalez, & Aramaki, 2010), TAGs (Binks & Marinopoulos, 2017; Mishima, Suzuki, Sato, & Ueno, 2016), fatty acids (Binks, Garvey, & Vieira, 2016), fatty alcohol (Fameau et al., 2015), and a combination of sucrose ester and lecithin (Patel, 2017d), as well as solid particles such as fluorinated particles (Binks, Johnston, Sekine, & Tyowua, 2015), have been reported to be capable of stabilizing air–oil interface in oil foams. Here, we mainly focus on edible oil foams stabilized by crystalline MAGs and DAGs. A recent work by Truong, Prakash, and Bhandari (2019) has applied a mixture of MAGs and phytosterols to produce oil foams from canola oil-based oleogels. The obtained samples had overrun percentages ranging from 47% to 52% and showed excellent storage stability that could maintain the initial foam volume as long as 2 months. It is believed that the foamability and foamstability of oil foams are determined by the phase behavior (solubility boundary) of crystalline stabilizers in the bulk oil (Binks et al., 2016; Chen, Van Damme, & Terentjev, 2009; Gunes et al., 2017; Shrestha et al., 2006b; Shrestha et al., 2006c; Shrestha, Aramaki, Kato, Takase, & Kunieda, 2006a), which can be controlled by altering types and concentrations of stabilizers, foaming temperature, storage temperature, and bulk oil types (Fameau & Saint-Jalmes, 2017). For example, when foaming temperature is well below the solubility boundary, MAGs or DAGs crystallize into particles to stabilize the air bubbles formed upon whipping. However, at a temperature above the solubility boundary, MAGs or DAGs are soluble in the bulk oil and no foams can be formed. Moreover, an increase in stabilizer concentration means that more particles can be absorbed to stabilize the air–oil interface, while the non-adsorbed particles can result in gelling of the continuous phase for enhanced stabilization of the air bubbles (Fameau & Saint-Jalmes, 2017). The obtained oil foams are usually considered to be thermally responsive since the melting and crystallization of the crystalline particles are reversible as a function of temperature, and therefore, the stabilization and de-stabilization of oil foams can be adjusted simply by changing temperature (Binks & Marinopoulos, 2017; Binks et al., 2016; Fameau et al., 2015).

It should be noted that the nature of the crystalline particles (for example, size, shape, and polymorphs) as well as the bulk and interfacial rheological behaviors of the stabilizer-oil systems or the formed oleogels should also be taken into account to understand the formation and stabilization mechanisms of oil foams (Fameau & Saint-Jalmes, 2017; Heymans, Tavernier, Dewettinck, & Meeren, 2017). For example, Heymans et al. (2018) have endeavored to relate the crystalline structure of oleogels formed by tuning tempering protocols to the foamability and foam stability of the resultant oil foams. Gunes et al. (2017) have studied the interfacial contribution of crystalline particles to the foam stability in a MAG-high oleic sunflower oil system, where the adsorbed MAG crystals were found to form a jammed, closely packed layer that could stabilize the air bubbles against a dilution step.

3.5 | Oxidative stability

Lipid oxidation is one of the major factors accounting for food quality deterioration, during which free radicals, hydroperoxides and their decomposed compounds, and polymers can be formed, thus leading to loss of shelf-life and nutrition as well as formation of off-flavors and even harmful substances (Arab-Tehrany et al., 2012; Shahidi & Zhong, 2010). Various intrinsic and extrinsic factors can affect the oxidative stability of fats and oils, such as unsaturation of fatty acids and their positional distribution, minor components (for example, FFAs, MAGs, DAGs), metal ions, light, heat, enzymes, and antioxidants (Shahidi & Zhong, 2010) as well as physical states of lipids (Berton-Carabin, Ropers, & Genot, 2014; Decker et al., 2017; Villeneuve, Durand, & Decker, 2018; Waraho, Mcclements, & Decker, 2011). In terms of SLs, changes in fatty acid composition, positional distribution, and acylglycerol composition (for example, DAGs), as well as applied production and purification process, may bring about changes in their oxidative stability, which need to be systematically assessed before their applications in the food industry.

3.5.1 | Fatty acid profiles

Generally, an increase in the unsaturation of fatty acids results in decreased oxidative stability. PUFAs (for example, *n*-3 series) are usually incorporated into SLs to enhance nutrition (He et al., 2018; He et al., 2017b; Palacios, Ortega, Rubio-Rodríguez, & Busto, 2019; Wang, Wang, Wang, Jin, & Wang, 2018a), but these types of fatty acids are highly liable molecules and are easily susceptible to oxidation (Shahidi & Zhong, 2010). In contrast, a decreased unsaturated level leads to increased oxidative stability. As have mentioned above, vegetable oils such as high-oleic peanut oils (Martín, Grosso, Nepote, & Grosso, 2018; Nepote, Olmedo, Mestrallet, & Grosso, 2009), sunflower oils (Roman, Heyd, Broyart, Castillo, & Maillard, 2013; Smith, King, & Min, 2007), rapeseed oils (Merrill, Pike, Ogden, & Dunn, 2008; Petersen,

Kleeberg, Jahreis, Busch-Stockfisch, & Fritsche, 2012), and soybean oils (Napolitano, Ye, & Cruz-Hernandez, 2018) have been reported to exhibit a higher oxidative stability compared to their normal oils. The positional distribution of fatty acids on the glycerol backbone also affects their oxidative stability. A series of work in Wijesundera's group has found that TAGs with PUFAs at the *sn*-2 position were more stable against oxidation than those with PUFAs at the *sn*-1(3) position in bulk oils (Wijesundera, 2008; Wijesundera et al., 2008) and O/W emulsions stabilized by Tween 40 (Shen & Wijesundera, 2009). They have also pointed out that, however, further studies are still needed to verify these findings by changing TAG species or by using different types of emulsifiers, and to clarify the mechanism behind them (Shen & Wijesundera, 2009).

3.5.2 | Acylglycerol composition

Variations in acylglycerol compositions, as in the case of MAGs and DAGs, may result in changes in oxidative stability. Previous studies have shown contradictory results when comparing the oxidative stability of DAG-rich oils and traditional TAG oils, since some researchers observed a decreased oxidative stability (Kristensen, Nielsen, Jacobsen, & Mu, 2006; Qi et al., 2015; Wang et al., 2010), whereas others reported a similar or even slightly increased oxidative stability for DAG-enriched oils (Shimizu, Moriwaki, Nishide, & Nakajima, 2004). It is not surprising since so many factors can affect the oxidative stability of lipids as have mentioned above. For example, Wang et al. (2010) observed a shorter oxidation induction time of DAG-rich oils compared to their TAG oils, which were obtained by partial hydrolysis followed by two-step molecular distillation. Based on chemical composition analysis (that is, acylglycerol composition, fatty acid composition, and antioxidant content), they attributed the reduced oxidative stability of DAG oils to multiple effects including less steric hindrance of DAG molecules, higher unsaturation level, and lower content of antioxidants of DAG oils as well as intense processing conditions (for example, high-temperature purification). Therefore, it is important to keep in mind that other variables should be maintained as the same when investigating the effect of one factor like acylglycerol composition. This has been taken into consideration in the work of Qi et al. (2015), where column chromatography was adopted to obtain purified DAGs and TAGs, respectively, so that the difference in their oxidative stability could only result from their different acylglycerol composition and fatty acid composition, which is a forward step to understand the effect of acylglycerol composition on lipid oxidation. Nevertheless, continuous efforts are still required before a definite conclusion can be reached. It is suggested future researches are performed in a model system with a single fatty acid composition and meanwhile other factors should be maintained as the same except for their acylglycerol composition, so as to obtain more convincing results.

3.5.3 | Physical state

Various SLs such as DAGs (Diao et al., 2016; Long et al., 2015; Shin, Lee, & Lee, 2016) and HMFs (Sproston & Akoh, 2016a; Zou & Akoh, 2015a, 2015b) can be applied in the form of O/W emulsions. Compared to bulk oil, there are three physical regions in an O/W emulsion, namely, oil phase, interfacial region, and aqueous phase, which make lipid oxidation behavior more complicated. An in-depth understanding of the effect of the three regions on oxidative stability can be found in several excellent review papers (Berton-Carabin et al., 2014; Berton-Carabin, Sagis, & Schroën, 2018; Decker et al., 2017; Villeneuve et al., 2018). We mainly focus on several approaches that have been explored to curb lipid oxidation in O/W emulsion systems based on the latest advances, mainly by strengthening the interfacial layer, inhibiting oxidation-promoting factors (for example, aqueous metal ions and oxygen), and adding interfacial antioxidants.

Interfacial films formed by biopolymer-based emulsifiers can serve as a physical barrier against oxygens and prooxidants in the aqueous phase (Berton-Carabin et al., 2014). Increased emulsion oxidative stability has been reported when interfacial films are strengthened by crosslinking via high-pressure homogenization or transglutaminase (Phoon, Paul, Burgner, San Martin-Gonzalez, & Narsimhan, 2014) and by forming multilayer films (Hermund et al., 2016; Lesmes, Sandra, Decker, & Mcclements, 2010). Moreover, the aqueous composition (for example, metal ions and oxygen) can be controlled to inhibit lipid oxidation. Metal chelators such as EDTA have been widely used to inhibit lipid oxidation (Alamed, Mcclements, & Decker, 2006; Lee & Decker, 2011; Nielsen, Petersen, Meyer, Timm-Heinrich, & Jacobsen, 2004), but an increasing demand for clean-label products has advanced recent researches on natural metal chelators (Todokoro, Fukuda, Matsumura, Irie, & Hata, 2016) and active packaging with metal-chelating properties (Lin & Goddard, 2018; Tian, Decker, Mcclements, & Goddard, 2014). Unabsorbed emulsifiers available in the aqueous phase are also believed to be able to inhibit oxidation (Berton-Carabin et al., 2014). For example, Gumus, Decker, and Mcclements (2017) have observed reduced oxidative stability of O/W emulsions when the unabsorbed protein emulsifiers (lentil, pea, faba bean proteins) were washed away by substituting the aqueous phase with buffer solutions. But the oxidative stability could be restored when the proteins were added back, mainly due to iron-binding capacity of the unabsorbed proteins. Addition of other types of biopolymers in O/W emulsions, which are not intended to be used as emulsifiers, have also been reported to exhibit antioxidant capacity such as casein (Chen et al., 2017a), citrus pectin (Celus et al., 2018),

and sodium alginate (Salvia-Trujillo, Decker, & Mcclements, 2016), to name a few. In addition, a recent work by Johnson, Inchingolo, and Decker (2018) has reported that oxygen content in a fish oil-in-water system could be reduced by ≥95% by using commercially accessible oxygen scavenging packaging, which significantly improved the emulsion oxidative stability. Furthermore, recent interests have been focused on designing interfacial antioxidants for improved antioxidative abilities (Freiría-Gándara, Losada-Barreiro, Paiva-Martins, & Bravo-Díaz, 2018; Yesiltas et al., 2019), in which antioxidants can be endowed with interfacial activity by physical or chemical interactions with emulsifiers (Mcclements & Decker, 2017).

4 | HEALTH ASPECTS

In addition to their physicochemical properties, healthy aspects of SLs are also of great importance since numerous health issues such as obesity and cardiovascular diseases can be, to some extent, controlled by adjusting dietary ingestion of lipids and lipid-based products. The health benefits of most conventional SLs such as MLCTs, HMFSs, 1,3-DAGs are mainly attributed to their unique metabolic characteristics arising from specific fatty acid composition and/or positional distribution (Gollaher & Bistrian, 2018; Phuah et al., 2015; Şahin-Yeşilçubuk & Akoh, 2017). In the case of *trans*-free plastic fats, they can serve as an alternative to hydrogenated vegetable oils to reduce TFA consumption (Wagh & Martini, 2017). In this part, we first give a brief introduction to the digestion and adsorption of TAGs to facilitate the understanding of the nutritional values of SLs. Our main interest is focused on two novel strategies for dealing with the above health-related issues. The first strategy involves modulation of lipid digestion through the addition of biopolymers (for example, proteins and polysaccharides), while the second one is related to biopolymer-based oleogelation of liquid oils for partial or complete replacement of *trans* and/or saturated fat.

4.1 | Nutrition of conventional SLs

The incorporation of PUFAs seems to be the most direct approach to obtain SLs with enhanced nutrition, since PUFAs (for example, *n*-3 and *n*-6 series) have been demonstrated to exhibit excellent physiological functions (Czernichow, Thomas, & Bruckert, 2010; Ma, Jiang, & Lai, 2016; Monteiro et al., 2014; Nicholson, Khademi, & Moghadasian, 2013). For example, in a recent work by Xia et al. (2019), healthy palm based structured lipids have been produced by simply introducing α-linolenic acid (ALA) or eicosapentaenoic acid (EPA), and under optimal conditions, the ALA or the EPA content can reach 27.1% and 30.9%, respectively. Meanwhile, it should be kept in mind that the susceptible chemical structure of PUFAs must be carefully preserved to maintain their health benefits and prevent the adverse effects caused by lipid oxidation (Arab-Tehrany et al., 2012). Furthermore, a basic knowledge of TAG digestion and absorption behavior helps to understand the nutritional properties of SLs. Based on a recent book chapter by Ferreira and Tonetto (2017b), we can gain a general picture of metabolic pathways of TAGs. When TAGs are ingested, they go through a consecutive digestive tract (that is, mouth, stomach, and small intestine) where various digestive products including FAs, DAGs, and 2-MAGs can be produced from lipolysis by lipases and colipases. It is recognized that short- and medium-chain fatty acids (MCFAs) are directly absorbed in the stomach and/or intestine and eventually reach the liver for providing energy. In contrast, the final products of long chain fatty acids (LCFAs, with more than 16 carbons) and 2-MAGs are absorbed by forming micelles with the help of bile salts and further re-esterified into new TAGs present in the form of chylomicrons, which constitutes the main route to resynthesize TAGs in human body (Yanai et al., 2007).

Bearing this in mind, it is rather easy to figure out the nutritional properties of certain SLs such as MLCTs, HMFSs, and 1,3-DAGs. For MLCTs (for example, MLM-type), MCFAs are released during digestion, which can be readily metabolized to provide energy. In the case of HMFSs, palmitic acids are mainly at the *sn*-2 position (~40% to 60%) while unsaturated FAs (for example, oleic acids) at the *sn*-1,3 positions, and this characteristic TAG composition is beneficial to the absorption of palmitic acid and calcium by blocking the formation of insoluble calcium soaps (Şahin-Yeşilçubuk & Akoh, 2017). Moreover, their nutritional value can be further improved when PUFAs (Abed, Zou, Ali, Jin, & Wang, 2017; Wang, Zou, Miu, Jin, & Wang, 2019) and/or MCFAs (Álvarez & Akoh, 2016) are incorporated. DAGs, especially 1,3-DAGs, have been reported to reduce serum TAG/cholesterol level, enhance β-oxidation, suppress fat accumulation, and modulate glucose metabolism (Lee et al., 2019; Lo et al., 2008; Phuah et al., 2015). The health effects of 1,3-DAGs are primarily due to their different acylglycerol structure and metabolic characteristics compared to TAGs (Yanai et al., 2007). Obviously, 1,3-DAGs are digested without the formation of 2-MAGs, and therefore, the re-synthesis of TAGs via 2-MAG pathway can be inhibited (Flickinger & Matsuo, 2005; Phuah et al., 2015).

4.2 | Modulation of lipid digestion

Recent interests have been focused on tuning lipid digestion through biopolymer-based food structuring (Corstens et al., 2017; Guo et al., 2017; Sarkar et al., 2019), among which O/W emulsions have been extensively studied as a model system since their chemical composition, structure and physical properties can be simply manipulated to provide in-depth information on lipid digestion affected by biopolymers (Mcclements, 2018). Lipid digestion is an interfacial process

relying on collaboration work of lipases, colipases, and bile salts at the interface of the emulsified lipid droplets (Wilde & Chu, 2011), which may be interfered by biopolymers that are used to stabilize emulsions.

Different stabilizers show distinct performance in delaying lipolysis, due to their various initial nature (for example, composition, structure, and physicochemical properties) as well as the complex environment of digestive tracts (Mcclements, 2018; Sarkar et al., 2019; Wilde & Chu, 2011). Natural proteins have been widely used as effective emulsifiers to obtain physically stable O/W emulsions, but they are less effective in delaying lipolysis upon consumption. Protein-based emulsifiers may undergo proteolysis when they travel through the gastric intestinal tract (GIT) and moreover, they can also be easily displaced by surface-active bile salts in the small intestine (Sarkar, Horne, & Singh, 2010; Sarkar, Ye, & Singh, 2016; Singh & Sarkar, 2011). In contrast, Pickering emulsions stabilized by various edible particles, such as cellulose nanocrystals (Bai et al., 2019; Liu, Kerr, & Kong, 2019) and cellulose nanofibers (Winuprasith et al., 2018), have been found to be better at delaying lipid digestion. For instance, Bai et al. (2019) constructed a corn oil-in-water emulsion stabilized by cellulose nanocrystals (CNC), which was subject to a simulated GIT model to assess lipid digestion behavior. The results showed that compared to a gum arabic-stabilized emulsion, the CNC-stabilized Pickering emulsion exhibited a slower initial rate of lipid digestion and finally fewer amounts of FFAs were released. This phenomenon could be explained by multiple effects: (a) inhibition of bile salt and lipase adsorption due to the irreversible adsorption of CNC to the lipid droplet; (b) a reduced available surface area for the bile salts and lipase to bind as a result of the coalescence and flocculation of the lipid droplets; and (c) suppressed lipolysis because of accumulation of FFAs at the lipid droplet surfaces (Bai et al., 2019). Furthermore, it has been found that the ability of natural proteins (for example, whey protein isolate) in delaying lipid digestion can be enhanced by coating with cellulose nanocrystals (Sarkar, Li, Cray, & Boxall, 2018). In Table 7, various types of biopolymers, addition amount, their effect and mechanism for delaying lipid digestion in different systems have been summarized based on the most recently published works.

Nevertheless, it should be noted that most of the above-mentioned researches were conducted using O/W emulsion as a model system and only *in vitro* GIT models were adopted to evaluate a rather simple lipid digestion behavior (that is, FFA release). In fact, lipid digestion and adsorption in a real food system under physiological conditions are much more complicated, which requires further in-depth investigations with more advanced methods. For example, most recently, Deloid et al. (2018) have demonstrated the delaying effect of fibrillar nanocellulose (NC) on TAG digestion and absorption by adopting a series of *in vitro* (a simulated GIT and a small intestinal epithelial model) and *in vivo* (a rat gavage model) approaches. They also explored the possible mechanisms through particle size analysis, SEM observation, and molecular dynamics simulation. Furthermore, it is indicated that the chemical structure of lipids (for example, fatty acid unsaturation and chain length) may affect lipid digestion in the presence of NC, but no definite conclusion could be made due to complex food matrix in their studies (Deloid et al., 2018). SLs can be produced with modified fatty acid composition and/or positional distribution, and we have also discussed their metabolic properties in relation to nutritional values. However, to the best of our knowledge, there are very few reports on the digestion and adsorption behavior of SLs in the presence of biopolymers. It can be anticipated that more future researches will focus on this new field especially when SLs are applied in the form of emulsions stabilized by various biopolymers, which may lay a basic foundation for understanding the lipid digestion behavior in more complex food systems.

4.3 | Oleogelation for reduced *trans*/saturated fats

Public concerns over consumption of saturated fats and/or *trans* fats as well as interesterified fats of palm origin can be addressed by adopting an oil structuring strategy, where liquid oils can be physically immobilized to obtain oleogels with solid-like functionality and improved lipid profile (that is, incorporation of more unsaturated fatty acids; Patel, 2017b; Patel & Dewettinck, 2016). This novel physically structured oil has been extensively explored in various food applications, such as reformulated meat products (Jimenez-Colmenero et al., 2015; Patel & Dewettinck, 2016), bakery products (Demirkesen & Mert, 2019; Patel & Dewettinck, 2016), margarines and spreads (Martins et al., 2018; Patel & Dewettinck, 2016), chocolate products (Patel & Dewettinck, 2016; Stortz & Marangoni, 2011), ice creams (Martins et al., 2018), and delivery systems for lipophilic bioactive molecules (O'sullivan, Barbut, & Marangoni, 2016). Most recently, SLs have been incorporated as the oil substrates to form oleogels with additional health benefits. For example, Willett and Akoh (2019a) have produced a novel oleogel using an MLM-type SL as the oil phase. In the following work, they have successfully explored the potential of SL-based oleogels as a substitute for shortening yellow cake production, and the textural properties of the obtained oleogel cake were similar to the shortening cake (Willett & Akoh, 2019b).

Oil structuring agents are believed to play a significant role in oleogelation, which can be primarily divided into two categories: low molecular weight oleogelators (LMWOs) and high molecular weight oleogelators. In the past decade, the fundamental understanding of LMWO-based oleogels has been substantially investigated (Co & Marangoni, 2012;

TABLE 7 Types of biopolymers, addition amount, their effect and mechanism for delaying lipid digestion in different systems

Biopolymer types	Addition amount	Oil system	Evaluating methods for lipid digestion	Effect of biopolymers on lipid digestion	Possible mechanism for delayed or inhibited lipid digestion	References
Fibrillar nanocellulose (CNF-50 nm and CNF-80 nm)	0.75 wt% for *in vitro* simulated GIT and *in vitro* small intestinal epithelial model tests, and 1.0 wt% for *in vivo* animal test	A high-fat model food (heavy cream) with or without 0.75 wt% CNF or CNC with a final fat concentration of 13.3 wt%; **Control**: the same high-fat model food with cellulose nanocrystals (CNC-25 nm) as well as micrometer-scale cellulose fibers (CMF) of the same origin, wheat dextrin, and psyllium husk dietary fibers; **Positive control**: the same high-fat model food with the chemical lipase inhibitor orlistat at 4.8 mg/mL	• FFA release by using an *in vitro* simulated GIT model • Fat adsorption by using an *in vitro* triculture small intestinal epithelial model; • Postprandial serum TAG level by applying an *in vivo* rat gavage model.	• CNF-50 nm at 0.75 wt% reduced FFA release by 48.4% quantified by pH-Stat titration, and by 40.1% assessed by fluorometric FFA assay. • The translocation of TAG and FFA across an *in vitro* small intestinal epithelial model was significantly reduced by the addition of 0.75 wt% CNF-50 nm. • Addition of 1.0 wt% CNF-50 nm decreased the postprandial rise in serum TAG 1 hr after gavage with the high-fat model food.	• Reducing the available surface area for lipase binding due to a coalescence of fat droplets on CNF • Impairing interfacial displacement of lipases at the lipid droplet surface and impairing solubilization of lipid digestion products, caused by sequestration of bile salts by CNF	Deloid et al. (2018)
Cellulose nanocrystals (CNC)	0.1 wt %–1.0 wt % of the aqueous phase	A corn oil-in-water Pickering emulsion stabilized by CNCs with an oil phase concentration of 10 wt %; **Control**: CNC suspensions without oils under the same conditions; **Reference**: a corn oil-in-water emulsion stabilized by gum Arabic (GA) with other conditions being the same	• FFA release in the small intestine stage over time	• The final released FFAs were reduced by ~40% in emulsions stabilized by 0.75 wt% CNC, compared to GA-stabilized emulsions where lipids were almost fully digested.	• Inhibiting bile salt and lipase adsorption because of the irreversible adsorption of CNC to the lipid droplet. • Reducing available surface area for the bile salts and lipase to bind as a result of the coalescence and flocculation of the lipid droplets. • Suppressing lipolysis due to accumulation of FFAs at the lipid droplet surfaces.	Bai et al. (2019)

(Continues)

TABLE 7 (Continued)

Biopolymer types	Addition amount	Oil system	Evaluating methods for lipid digestion	Effect of biopolymers on lipid digestion	Possible mechanism for delayed or inhibited lipid digestion	References
Cellulose nanofibrils (CNF) TEMPO-modified cellulose nanofibrils (TEMPO-CNF) Cellulose nanocrystals (CNC)	CNF (0.23 wt%, 1.17 wt%, and 2.34 wt% of the initial emulsion), TEMPO-CNF (0.12 wt%, 0.23 wt%, 0.59 wt%, and 0.85 wt%), and CNC (0.23 wt%, 1.17 wt%, 2.35 wt%, 4.70 wt%, and 7.04 wt%)	A mixture of a Tween-80 stabilized canola oil-in-water emulsion and nanocellulose suspensions with a final concentration of 2.2 wt% canola oil; **Control**: O/W emulsions without any addition of CNF, TEMPO-CNF or CNC; **Reference**: 2.34 wt% cellulose suspensions	• FFA release by using static *in vitro* digestive model consisting of salivary, gastric, and intestinal phases	• The initial lipolysis rate at high concentrations of NC (1 wt% CNF, 0.25 to 0.36 wt% TEMPO-CNF and 2 to 3 wt% CNC) was significantly reduced. Nonetheless, the final lipolysis extent was very similar for all the emulsions (47% to 55%).	• The delay of the initial lipolysis lies in the difference of the three nanocellulose at higher concentrations in adjusting viscosity of the system with CNC being more effective by forming a hydrogel during gastric and intestinal digestion, but the access of lipase to lipid droplets results in almost the same final lipolysis extent due to larger pore size of CNF or CNC.	Liu et al. (2019)
Nanofibrillated mangosteen cellulose (NFC)	0.10 wt% to 0.70 wt% NFC	A soybean oil-in-water emulsion containing 10 wt% of lipid phase (9.99 wt% soybean oil and 0.01 wt% vitamin D3) stabilized by NFC; **Reference**: O/W emulsions stabilized by whey protein isolate (WPI)	• FFA release (the initial digestion rate and the final extent of lipid digestion) by using a multi-stage static GIT model	• NFC-stabilized emulsions exhibited a slower initial rate and final extent of lipid digestion especially at higher NFC concentrations, compared to WPI-stabilized emulsions,	• NFC could act as a physical barrier at the lipid droplet surfaces, promote droplet flocculation in the gastric phase, and increase the viscosity of the aqueous phase.	Winuprasith et al. (2018)
Composite whey protein isolate-cellulose nanocrystals (WPI-CNCs)	1 wt% WPI and 1 or 3 wt% CNCs	A mixture of a sunflower oil-in-water emulsion stabilized by WPI and CNC dispersions with final concentration of 20 wt% oil, 1 wt% WPI and 1 or 3 wt% CNCs (referred to W1C1 or W1C3, respectively); **Control**: O/W emulsions stabilized by whey protein isolate (WPI) (referred to W1)	• FFA release in simulated duodenal conditions	• W1C1 showed a significant decrease in rate and extent of lipid digestion compared to W1, and a further decrease of lipid digestion was observed with increasing CNC concentration to 3 wt% in W1C3.	• The interfacial displacement and solubilization of digestive products by bile salts were restricted due to the binding of bile salts with CNCs. • The overall surface area for lipid digestion was reduced due to lipid droplet bridging induced by CNCs present in the continuous phase.	Sarkar et al. (2018)

TABLE 8 Summary of selected examples of biopolymer-based oleogels regarding oleogelation strategy, type of oleogelator, oil type, heating or drying method, and food application

Oleogelation strategy	Type of biopolymer-based oleogelator	Oil Type	Heating/drying method	Food application	Reference
Direct dispersion	EC (20 cP) with soy lecithin	high oleic canola oil:	Heating at 140 °C for 40 min	—	Aguilar-Zarate, Macias-Rodriguez, Toro-Vazquez, and Marangoni (2019)
	EC (10 cP) with SMS	Canola oil	Heating to 140 °C held for 10 min	Frankfurters	Barbut, Wood, and Marangoni (2016c)
	EC (10 cP) with SMS	Canola oil	Heating to 140 °C held for 10 min	Breakfast sausage	Barbut, Wood, and Marangoni (2016b)
	EC (10 cP) with SMS	Canola oil	Heating to 140 °C held for 10 min	Comminuted meat products	Barbut, Wood, and Marangoni (2016a)
	EC (10 cP) with SMS	Beef fat, rendered beef fat, canola oil, soybean oil, or flaxseed oil	Heating to 140 °C held for 10 min	Comminuted meat products	Barbut and Marangoni (2019)
	EC (20 cP)	Canola oil	Heating to 140 °C held for 10 min	Pork liver pâté	Barbut, Marangoni, Thode, and Tiensa (2019)
	EC (10 cP) with SMS	A mixture of olive, linseed, and fish oils	Heating to 160 °C held for 10 min	Pork liver pâté	Gomez-Estaca et al. (2019a)
	EC (10 cP) with SMS	A mixture of olive, linseed, and fish oils	Heating to 160 °C held for 10 min	Pork burgers	Gomez-Estaca, Pintado, Jimenez-Colmenero, and Cofrades (2019b)
	EC (10 cP, 45 cP, and 100 cP)	Cinnamon essential oil	Heating to 145 °C (EC, 10 cP), 165 °C (EC, 45 cP), 180 °C (EC, 100 cP)	Antibacterial edible packaging	Zhang et al. (2019)
	EC (45 cP)	Extra virgin olive oil	Heating to 140 °C held for 10 min	—	Giacintucci et al. (2018)
	EC polymers EC7 (6~9 cP), EC20 (18~22 cP), EC50 (45~55 cP), EC100 (90~110 cP), or EC200 (180~220 cP)	Soybean oil	Heating to 130 °C until fully dissolved	Wheat breads	Ye, Li, Lo, Fu, and Cao (2019)

(Continues)

TABLE 8 (Continued)

Oleogelation strategy	Type of biopolymer-based oleogelator	Oil Type	Heating/drying method	Food application	Reference
	EC (10 cP, 20 cP, 45 cP)	Canola oil	Heating to 150 °C until fully dispersed	Lipophilic bioactive delivery (that is, beta-carotene)	O'sullivan et al. (2017)
	EC (22 cP) with *Kokum* fat and SMS	Flaxseed oil, rice bran oil, or olive oil	Heating to 135 to 140 °C until dissolved	—	Nagavekar, Kumar, Dubey, and Singhal (2019)
	EC (10 cP, 20 cP) with distilled MAG	High oleic sunflower oil	Heating to 180 °C held for 10 min	Ice cream formulations	Munk, Utoft, Larsen, Needham, and Risbo (2019)
	EC (45 cP) with stearyl alcohol and stearic acid	Canola oil	Heating to 150 °C held for 20 min	—	Gravelle, Blach, Weiss, Barbut, and Marangoni (2017); Gravelle, Davidovich-Pinhas, Barbut, and Marangoni (2017)
	EC (45 cP) with either oleic acid or oleyl alcohol	Canola oil, soybean oil, flaxseed oil, or castor oil	Heating to 150 °C held for 20 min	—	Gravelle, Davidovich-Pinhas, Zetzl, Barbut, and Marangoni (2016)
	Hydrophobically modified chitin	Sunflower oil	Heating at 60 °C overnight	—	Huang et al. (2015)
	Crude chitin with surfactants (that is, PC, modified PC, and SMS), or chitin nanocrystal with PC	Sunflower oil	Heating at 85 °C for 30 min	—	Nikiforidis and Scholten (2015)
	MC coated palm stearine microcapsule	Sunflower oil	Heating at 70 °C to melt the microcapsule	—	Patel (2017c)
	Dried whey protein isolate aggregates	Sunflower oil	Freeze drying with high freezing rate	—	De Vries et al. (2017a)
Emulsion-based template	HPMC with XG, or MC with XG	Sunflower oil	Oven drying (50, 60, 70, and 80 °C)	—	Patel et al. (2014b)
	MC with XG	Sunflower oil	Oven drying (50, 60, 70, and 80 °C)	Classic 4/4 sponge cakes	Patel et al. (2014a)
	GLT with XG	Sunflower oil	Oven drying (70 °C, 48 hr), or freeze drying (72 hr)	—	Patel et al. (2015)
	Zein particle/stearate complexes	Soybean oil	Freeze drying (over 24 hr)	—	Gao et al. (2014)

(Continues)

TABLE 8 (Continued)

Oleogelation strategy	Type of biopolymer-based oleogelator	Oil Type	Heating/drying method	Food application	Reference
	GLT/tannic acid/flaxseed gum nanocomplexes	Soybean oil	Oven drying (60 °C, 48hr), or freeze-drying (48 hr)	—	Qiu et al. (2018)
	Soybean protein, or soybean protein/κ-carrageenan complexes	Sunflower oil	Oven drying (60 °C)	—	Tavernier et al. (2017)
	SC/SA mixtures, or SC/SA conjugates	Sunflower oil	Oven drying (35 °C, 24 hr)	—	Wijaya et al. (2019)
	Regenerated cellulose with CMC	Sunflower oil	Freeze drying (48 hr)	—	Jiang et al. (2018)
	Citrus pectin with tea polyphenol-palmitate particles	Camellia oil	Freeze drying (48 hr)	Classic 4/4 sponge cakes	Luo et al. (2019)
	HPMC with various thickening agents (that is, CMC, XG, SA, GG, arabic gum, flaxseed gum, or locust bean gum)	Soybean oil	Vacuum oven drying (90 °C, ~12 hr)	—	Meng et al. (2018a)
	HPMC with XG; or MC with XG	Soybean oil	Vacuum oven drying (90 °C, ~12 hr)	—	Meng et al. (2018c)
	HPMC with XG	Soybean oil	Vacuum oven drying (90 °C, ~12 hr)	—	Meng, Qi, Guo, Wang, and Liu (2018b)
	SC/ XG/GG ternary mixtures, SC/XG or SC/GG binary mixtures	Sunflower oil	Oven drying (70 °C), or freeze drying	—	Abdolnnaleki, Alizadeh, Nayebzadeh, Hosseini, and Shahin (2019)
Foam-based template	HPMC	Sunflower oil	Freeze drying	Muffins	Oh and Lee (2018)
	HPMC	Canola oil	Freeze drying	Meat patties	Oh et al. (2019)
	HPMC	Sunflower oil	Freeze-drying	Classic 4/4 sponge cakes	Patel and Dewettinck (2015a)
	HPMC	Sunflower oil	Freeze drying	—	Patel, Schatteman, Lesaffer, and Dewettinck (2013)
	HPMC	Canola oil	Freeze drying	Sandwich cookie creams	Tanti et al. (2016a)
	HPMC, or MC	Peanut oil	Freeze drying	Peanut butter	Tanti et al. (2016b)
	GLT with XG	Canola oil	Freeze drying (24 hr)	—	Abdollahi et al. (2019)

Abbreviations: EC, ethyl cellulose; SMS, sorbitan monostearate; MAG, monoacylglycerol; PC, phosphatidylcholine; MC, methyl cellulose; HPMC, Hydroxypropyl methylcellulose; XG, xanthan gum; GLT, gelatin; SC, sodium caseinate; SA, sodium alginate; CMC, carboxymethyl cellulose; GG, guar gum.

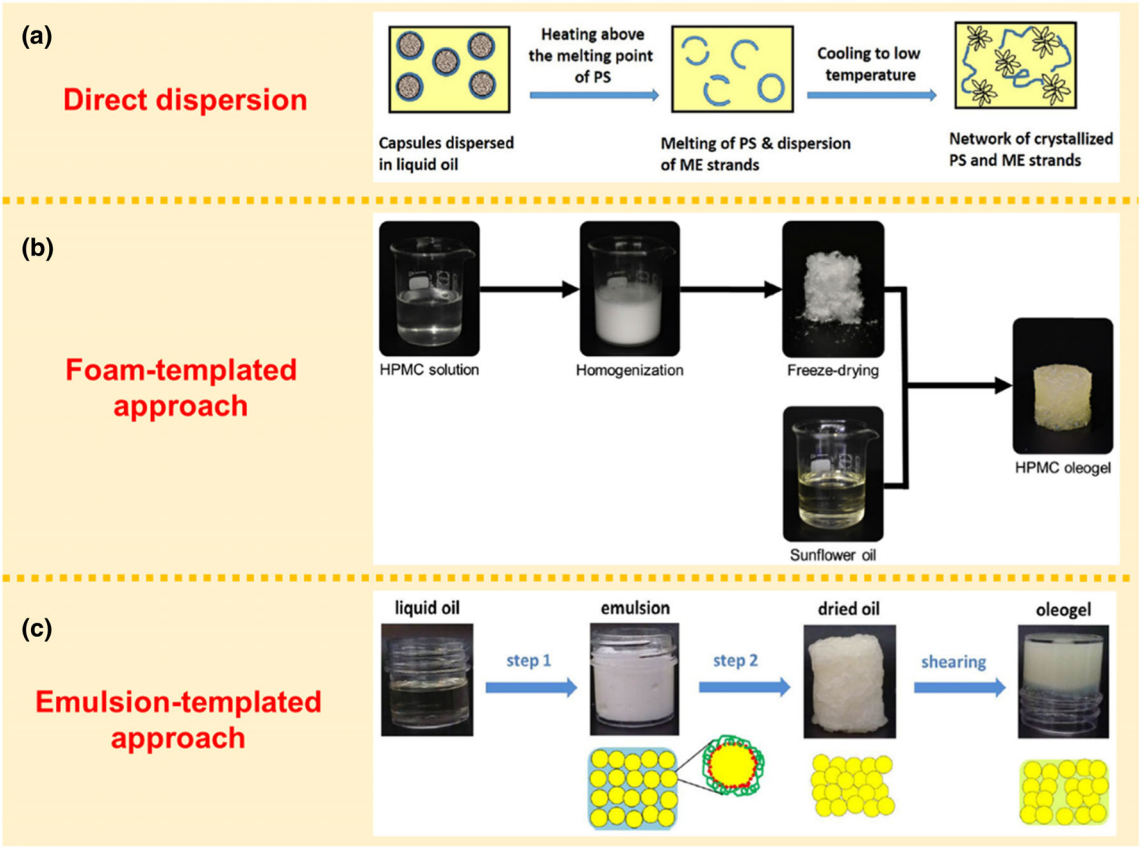

FIGURE 4 Schematic illustration of selected examples of different biopolymer-based oleogelation strategies. (a) Oleogels structured with methylcellulose coated palm stearine (PS) capsules using a direct dispersion method (Patel, 2017c). (b) Oleogels structured with freeze-dried hydroxypropyl methylcellulose (HPMC) by adopting a foam-templated approach (Oh & Lee, 2018). (c) Oleogels formed by using water continuous emulsions stabilized by a combination of gelatin and xanthan gum as the templates (Patel et al., 2015)

Dassanayake, Kodali, & Ueno, 2011; Doan, Tavernier, Okuro, & Dewettinck, 2018; Patel, 2017a). Recently, oil structuring with biopolymers (for example, proteins and polysaccharides) has aroused considerable research interest, since most of them are food-grade macronutrients, commercially available and relatively inexpensive (Davidovich-Pinhas, 2019; Davidovich-Pinhas, Barbut, & Marangoni, 2016; Patel, 2018; Scholten, 2019). In this regard, we mainly concentrate on the most recent progress of biopolymer-based oleogels and their food applications. Generally, several strategies including direct dispersion of structuring agents, indirect methods (for example, an emulsion- or a foam-templated approach), structured emulsions and oil bulking systems can be applied to physically structure oils (Jimenez-Colmenero et al., 2015; Patel & Dewettinck, 2016; Singh et al., 2017). It should be noted that we mainly focus on direct dispersion, emulsion-, or foam-templated approaches (Figure 4), without considering the incorporation of structured emulsions and oil bulking systems since these two systems, to put it strictly, cannot be regarded as oleogels due to the presence of water. Table 8 presents some latest reports of biopolymer-based oleogels in terms of oleogelation strategy, type of oleogelator, oil type, heating or drying method, and food application.

Ethylcellulose (EC), a widely used food-grade polymer, has been extensively explored by Marangoni's group to structure oils in various food formulations (Co & Marangoni, 2012; Davidovich-Pinhas et al., 2016; Gravelle & Marangoni, 2018). EC has attained the Generally Recognized as Safe (GRAS) status by the US Food and Drug Administration for direct food use (21 Code of Federal Regulations section: 172.868). Like LMWOs, EC can be directly dispersed in the oil phase, and the formation of EC-oleogels is believed to involve the dissolution of the polymer in the oil above the glass transition temperature (~140 °C) and subsequent hydrogen bonding between EC molecules after cooling below the gelling point (Davidovich-Pinhas, Barbut, & Marangoni, 2015). The mechanical properties of the EC-oleogels can be altered by simply changing solvent polarity, EC type (for example, molecular weight, substitution degree, and concentration), and processing conditions (for example, setting temperature and surfactant addition; Davidovich-Pinhas et al., 2016). Most recently, novel applications of EC-oleogels have been exploited as the oil phase to construct cinnamon oil-loaded O/W emulsions with antibacterial properties (Zhang et al., 2019), or as the delivery system of lipophilic bioactive (for example, β-carotene) for controlled transfer and enhanced

stability (O'sullivan, Davidovich-Pinhas, Wright, Barbut, & Marangoni, 2017). Other structuring agents, such as hydrophobically modified chitin (Huang et al., 2015), crude chitin or chitin nanocrystal with surfactants (that is, phosphatidylcholine (PC), modified PC, or sorbitan monostearate) (Nikiforidis & Scholten, 2015), and palm stearine microcapsule coated with methylcellulose (Patel, 2017c), have also been reported to produce oleogels using a direct dispersion method. Furthermore, submicron heat-set whey protein aggregates (WPAs) have been demonstrated to be dispersed and form a network in vegetable oils (De Vries, Wesseling, Van Der Linden, & Scholten, 2017c). Oil polarity has been found to affect the mechanical strength of the WPA-stabilized oleogels, and an increase in oil polarity produces a weaker gel due to enhanced protein-solvent interactions (De Vries, Gomez, Van Der Linden, & Scholten, 2017b). In addition, the gel strength can be enhanced by water addition and subsequent heat treatment, which may promote protein–protein interactions (De Vries, Jansen, Van Der Linden, & Scholten, 2018). In the above-mentioned researches, the dispersion of WPAs has to be aided by using a solvent exchange procedure to prevent agglomeration of protein particles, while the directly freeze-dried products are not able to form an oleogel (De Vries et al., 2017b). However, they have recently observed that freeze-dried products also show potential for oleogelation by modifying drying process, where dried protein aggregates from a low polarity solvent or with higher freezing rate before drying have been found to be effective to gel a liquid oil (De Vries, Gomez, Jansen, Van Der Linden, & Scholten, 2017a).

In contrast, an emulsion-templated or a foam-templated approach for oil structuring is considered to be an indirect method to obtain oleogels by first constructing a water continuous emulsion or an aqueous foam using biopolymers as stabilizers followed by removing water and further shearing (for dried emulsion) or mixing with liquid oils (for dried foams; Patel & Dewettinck, 2016). To form an emulsion-templated oleogel, the drying process is required for the removal of water in the continuous phase, but it has been found that oil droplets in concentrated emulsions tend to coalesce during drying (Patel et al., 2014b). A strengthened emulsion interface has been reported to be effective against oil droplet coalescence by utilizing a synergistic effect of biopolymers, such as a combination of regenerated cellulose and carboxymethyl cellulose (CMC; Jiang et al., 2018), methylcellulose (MC)/hydroxypropyl methylcellulose (HPMC) and xanthan gum (Meng, Qi, Guo, Wang, & Liu, 2018a, 2018b, 2018c; Patel, Cludts, Bin Sintang, Lesaffer, & Dewettinck, 2014a), and gelatin and xanthan gum (XG) (Patel et al., 2015). In a recent work, a ternary complex formed by gelatin, tannic acid, and flaxseed gum has also been fabricated to produce a stable oleogel from soybean oil-in-water emulsions, due to stabilizing effect of both the interfacial adsorbed particles and the bulk polymer gel network (Qiu, Huang, Li, Ma, & Wang, 2018). To produce oleogels in a controlled manner, it is also important to understand the relationship between emulsion stability and oleogel strength. Meng, Qi, Guo, Wang, and Liu (2018a, 2018b) have reported a positive correlation between them, that is, the mechanical strength and oil binding capacity can be improved by using a more stable emulsion as the template. However, an opposite phenomenon has been observed in another work (Tavernier, Patel, Van Der Meeren, & Dewettinck, 2017), where a less stable protein stabilized emulsion produced a firmer oleogel compared to a less firm oleogel structured with protein/κ-carrageenan complexes from a more stable emulsion. They have attributed this result to the suboptimal protein interfacial conformation after complexation with the polysaccharide. Recently, the oxidative stability of emulsion-templated oleogels has also aroused researcher's attention especially when PUFA-rich oils are incorporated. For example, it has been reported that oleogels of camellia oil (rich in oleic acids) structured with tea polyphenol-palmitate particles and citrus pectin showed higher oxidative stability compared to the liquid oil, probably as a result of both the formed gel structure and antioxidant properties of tea polyphenol-palmitate particles (Luo et al., 2019). However, it should be noted that the oleogel in this study was obtained from freeze-dried emulsions, and oven heating can be more suitable for industrial production of oleogels. In a recent work of Meng, Qi, Guo, Wang, and Liu (2018c), soybean oil in water emulsions stabilized by HPMC/XG or MC/XG have been used as the template to produce oleogel by applying vacuum (90 °C, ~12 hr) or normal oven drying (90 °C, ~48 hr). It has been found that oleogels showed better oxidative stability than soybean oil during storage (20 °C, 20 days). Moreover, vacuum drying was better at delaying oxidation than normal oven drying.

In terms of a foam-templated approach, a porous material is usually constructed from dried aqueous foams and subsequently mixed with liquid oils to produce oleogels, which have been applied in muffins (Oh & Lee, 2018), sandwich cookie creams (Tanti, Barbut, & Marangoni, 2016a), meat patties (Oh, Lee, Lee, & Lee, 2019), and peanut butter (Tanti, Barbut, & Marangoni, 2016b). HPMC or MC has been the most widely used structuring agent to develop foam-templated oleogels (Oh & Lee, 2018; Oh et al., 2019; Tanti et al., 2016a, 2016b). Recently, a strong oleogel stabilized by a combination of gelatin and xanthan gum has also been obtained, which can maintain its structure even at high temperatures (<100 °C) (Abdollahi, Goli, & Soltanizadeh, 2019). In addition to aqueous foams, a κ-carrageenan hydrogel has also been dried to obtain a porous structure (aerogel) as a template to produce oleogels (Plazzotta, Calligaris, & Manzocco, 2019). The drying method significantly affects the physicochemical properties of the resultant oleogels. It has been found that supercritical-CO_2-drying resulted in a hard and shrunk oleogel with lower oil content (80%) and

no oil release, whereas freeze-drying produced a soft oleogel with higher oil content (97%) and oil release (49%; Plazzotta et al., 2019). Compared to the direct dispersion method using EC or the emulsion-templated approach, the foam-templated method seems to have little or no impact on oil quality since the drying process is conducted in the absence of oil. For instance, foam-templated oleogels structured with HPMC (Oh et al., 2019) or gelatin with XG (Abdollahi et al., 2019) have showed greater oxidative stability than unstructured oils. Therefore, foam-templated oleogels may exhibit great potential in food applications where PUFA-rich oils or SLs are incorporated for improved lipid profile.

Based on what has been discussed above, it can be anticipated that future research work will focus on the exploration of other various types of biopolymers such as hydrophobically modified polysaccharides and commercially available emulsifiers or foaming agents to produce oleogels by adopting different oil structuring strategies. Moreover, different kinds of SLs with health benefits can be introduced to oleogels to further improve their nutrition. More extensive studies are also required to assess the performance and applicability of oleogels, especially SL-based oleogels, in broader food applications. Furthermore, considering the more unsaturated lipid profile of oleogels, it is important to further evaluate their oxidative stability during oleogelation where various heating/drying conditions may be applied (Table 8) and in real processed foods as affected by extra processing steps and other food components. In addition to the technological feasibility of oleogels for reducing *trans* and/or saturated fats in various food formulations, regulatory approvals and prescribed conditions are still required before they are practically adopted in commercial food products in different regions, such as the products that oleogels can be used in, and the type and maximum usage amount of oleogelators. Fortunately, most biopolymers explored in published works (Table 8) have been directly or indirectly used as GRAS food additives, including xanthan gum (172.695), acacia gum (gum arabic) (172.780), hydroxypropyl methylcellulose (172.874), methylcellulose (182.1480), guar gum (184.1339), locust bean gum (184.1343), sodium alginate (184.1724), and sodium caseinate (182.1748), to name a few. Therefore, in future studies, the current regulatory restrictions should also be considered when developing biopolymer-based oleogels and relevant food products, which may in turn provide basic technological guidance for probable future regulations governing real food applications of oleogels.

5 | POTENTIAL FOOD APPLICATIONS OF SLS

Conventional SLs are usually obtained with desirable physical properties and/or nutritional values, among which some of them (for example, *trans*-fee plastic fats, CBEs, HMFSs, and MAGs/DAGs) are synthesized mainly for specific purposes, while others such as MLCTs, DAG-rich oils, and PUFA-enriched SLs are produced with various potential food applications. For example, *trans*-fee plastic fats are mainly used as shortening and margarine fats to replace traditionally used hydrogenated oils and animal fats. Extensive studies have been conducted to exploit novel *trans*-free fats and assess their potential performance in various food products such as margarines (Hu, Xu, & Yu, 2017; Lakum & Sonwai, 2018; Li et al., 2018b; Ornla-Ied, Sonwai, & Lertthirasuntorn, 2016; Pande, Akoh, & Shewfelt, 2012), shortenings (Saghafi et al., 2018; Xu et al., 2018), as well as fat-frozen special fats (Zhu et al., 2019; Zhu et al., 2018b; Zhu et al., 2017). CBEs, as the name indicates, have been primarily applied to partially substitute cocoa butter in chocolate products, such as dark chocolates (De Clercq et al., 2017; Jin, Jin, Wang, & Akoh, 2019; Souza & Block, 2018), white chocolates (Bahari & Akoh, 2018b), and milk chocolates (Ackar et al., 2015). Similarly, HMFSs particularly those enriched in PUFAs has been widely applied in emulsion-based infant formula (Li, Sabir, Baeshen, & Akoh, 2015b; Zou & Akoh, 2015a, 2015b). MLM-type SLs have been explored in various food-related systems, such as cooking oil for sweet potato chips, energy bars, beverages, and edible films (Utama et al., 2019). MAGs/DAGs have been largely used as emulsifiers for a long time in many food products including baking products, margarines, and frozen desserts (Norn, 2015). DAG-rich oils have shown great potential in numerous food applications, such as frying shortening, stabilizer, or potential antioxidant in emulsions, and even pharmaceutical drugs (Lee et al., 2019). DAG oils used to be commercially available as a functional oil due to their potential health benefits particularly of 1,3-DAGs. Unfortunately, they have not been sold on the market since 2009 because of the discovery of harmful processing contaminants (that is, glycidols esters) in the products, which remains unsolved until nowadays and requires continuous researches.

It should be pointed out that the increasing research interest in novel lipid-based products such as oil foams and oleogels has provided a brand-new direction for exploiting SL-based food applications. It can be expected that SLs with nutritional values are incorporated into oil foams and oleogels as the oil phase, which can be further applied in various food products as discussed above.

6 | CONCLUDING REMARKS AND FUTURE TRENDS

SLs with specific physical properties and/or improved nutritional values can be obtained by using enzymatic modification, which can alter their fatty acid profiles and/or acylglycerol composition in a flexible manner. Particularly,

large-scale production of SLs can be promising by using immobilized lipase due to its advantages such as enhanced processing stability, easy recovery and reusability as well as suitability for continuous process. However, the relatively high production cost has been a major obstacle to hinder its wider applications in the food industry. It is anticipated that an inexpensive but facile immobilized lipase can be obtained by selecting cheaper soluble lipases, adopting rather simple immobilization strategy (that is, physical methods), and designing novel carriers with low cost but good function, which largely depends on the development and progress of both material science and enzyme engineering. It is also important to select suitable starting materials for the synthesis of desirable SLs. Fortunately, nature and modern biological techniques have enabled us to exploit and develop SLs based on novel fats and oils of diverse origins. For example, recent interest has been focused on producing PUFA-enriched SLs from algae, which can be a cheap, safe, and sustainable source of PUFAs in contrast to marine fish oil.

The physicochemical properties of SLs are largely defined by their chemical structures, molecular architectures, and are also affected by processing conditions, which in turn determine their applications in the food industry. Similar to conventional fats and oils, SLs can be used as important ingredients in various lipid-based products. Therefore, it is essential to assess their performance in a real food system, which may be influenced by the extra food processing process and interactions with other food components. Novel lipid-based products such as oil foams and oleogels have aroused considerable research interest in recent years, providing a new direction for developing healthy low-calorie SL-based products. In this regard, extensive researches are needed to further clarify the processing-structure-function relationship of SLs in various food products.

The nutritional values of SLs are primarily reflected by their unique metabolic properties from specific fatty acid profiles (that is, composition and/or positional distribution), the health benefits of PUFAs that are incorporated, as well as the reduced TFA ingestion in the case of *trans*-free plastic fats. Moreover, lipid digestion and absorption can be delayed by the addition of some types of biopolymers, which is beneficial to increase satiety and decrease lipid ingestion. In addition, oleogelation of liquid oils is believed to be a novel strategy to reduce *trans* and saturated fats and improve lipid profiles especially when SLs are used as the oil phase. However, information on the digestion and absorption behavior of various SLs as affected by their chemical structure (for example, fatty acid chain length and unsaturation level) in the presence of biopolymers is far from clear, and further in-depth work should be conducted to elucidate their metabolic process and mechanism to maximize their health aspects. Similarly, oleogels structured with biopolymers are also in their infancy; extensive future researches are still required to have an in-depth understanding of biopolymer-based oleogelation and explore their food applications in a wider range. Last but not least, we expect that the incorporation and discussion of olegels in the present work may extend our understanding of the concept of structured lipids to a broader sense, where novel physically structured oils could also be included in addition to the conventional SLs with modified chemical structures.

ACKNOWLEDGMENTS

This work was supported by the National Key Research and Development Plan (2016YFD0400200).

AUTHOR CONTRIBUTIONS

Yalong Guo was responsible for the compilation of the data, the writing of the draft, and the design of the tables. Zhixiang Cai and Yanping Xie previously produced an intern unpublished report on the functionality of SLs. Aiqin Ma contributed to the health-related considerations of SLs. Hongbin Zhang was responsible for the structure and the content of the manuscript, the correction, and the final approval for the version to be submitted. Pingfan Rao and Qiang Wang collaborated throughout the study and provided critical suggestions.

CONFLICTS OF INTEREST

The authors declare no conflict of interest.

ORCID

Hongbin Zhang https://orcid.org/0000-0002-4419-4818

LITERATURE CITED

Abdollahi, M., Goli, S. a. H., & Soltanizadeh, N. (2019). Physicochemical properties of foam-templated oleogel based on gelatin and xanthan gum. *European Journal of Lipid Science and Technology*, 1900196. https://doi.org/10.1002/ejlt.201900196

Abdolnnaleki, K., Alizadeh, L., Nayebzadeh, K., Hosseini, S. M., & Shahin, R. (2019). Oleogel production based on binary and ternary mixtures of sodium caseinate, xanthan gum, and guar gum: Optimization of hydrocolloids concentration and drying method. *Journal of Texture Studies*, https://doi.org/10.1111/jtxs.12469

Abed, S. M., Wei, W., Ali, A. H., Korma, S. A., Mousa, A. H., Hassan, H. M., … Wang, X. (2018). Synthesis of structured lipids enriched with medium-chain fatty acids via solvent-free acidolysis of microbial oil catalyzed by *Rhizomucor miehei* lipase. *LWT-Food Science and Technology*, *93*, 306–315. https://doi.org/10.1016/j.lwt.2018.03.057

Abed, S. M., Zou, X., Ali, A. H., Jin, Q., & Wang, X. (2017). Synthesis of 1,3-dioleoyl-2-arachidonoylglycerol-rich structured lipids by lipase-catalyzed acidolysis of microbial oil from Mortierella alpina. *Bioresource Technology*, *243*, 448–456. https://doi.org/10.1016/j.biortech.2017.06.090

Acevedo, N. C., & Marangoni, A. G. (2015). Nanostructured fat crystal systems. *Annual Review of Food Science and Technology*, *6*, 71–96. https://doi.org/10.1146/annurev-food-030713-092400

Acevedo, N. C., Peyronel, F., & Marangoni, A. G. (2011). Nanoscale structure intercrystalline interactions in fat crystal networks. *Current Opinion in Colloid and Interface Science*, *16*(5), 374–383. https://doi.org/10.1016/j.cocis.2011.05.004

Ackar, D., Skrabal, S., Subaric, D., Babic, J., Milicevic, B., & Jozinovic, A. (2015). Rheological properties of milk chocolates as influenced by milk powder type, emulsifier, and cocoa butter equivalent additions. *International Journal of Food Properties*, *18*(7), 1568–1574. https://doi.org/10.1080/10942912.2014.923440

Agregan, R., Barba, F. J., Gavahian, M., Franco, D., Khaneghah, A. M., Carballo, J., … Lorenzo, J. M. (2019). Fucus vesiculosus extracts as natural antioxidants for improvement of physicochemical properties and shelf life of pork patties formulated with oleogels. *Journal of the Science of Food and Agriculture*, *99*(10), 4561–4570. https://doi.org/10.1002/jsfa.9694

Aguilar-Zarate, M., Macias-Rodriguez, B. A., Toro-Vazquez, J. F., & Marangoni, A. G. (2019). Engineering rheological properties of edible oleogels with ethylcellulose and lecithin. *Carbohydrate Polymers*, *205*, 98–105. https://doi.org/10.1016/j.carbpol.2018.10.032

Akoh, C. C. (2017). *Food lipids: Chemistry, nutrition, and biotechnology*. Boca Raton, FL: CRC press.

Alamed, J., Mcclements, D. J., & Decker, E. A. (2006). Influence of heat processing and calcium ions on the ability of EDTA to inhibit lipid oxidation in oil-in-water emulsions containing omega-3 fatty acids. *Food Chemistry*, *95*(4), 585–590. https://doi.org/10.1016/j.foodchem.2005.01.041

Alfutimie, A., Al-Janabi, N., Curtis, R., & Tiddy, G. J. (2016). The effect of monoglycerides on the crystallisation of triglyceride. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *494*, 170–179. https://doi.org/10.1016/j.colsurfa.2016.01.029

Álvarez, C. A., & Akoh, C. C. (2016). Preparation of infant formula fat analog containing capric acid and enriched with DHA and ARA at the *sn*-2 position. *Journal of the American Oil Chemists' Society*, *93*(4), 531–542. https://doi.org/10.1007/s11746-016-2788-7

Arab-Tehrany, E., Jacquot, M., Gaiani, C., Imran, M., Desobry, S., & Linder, M. (2012). Beneficial effects and oxidative stability of omega-3 long-chain polyunsaturated fatty acids. *Trends in Food Science and Technology*, *25*(1), 24–33. https://doi.org/10.1016/j.tifs.2011.12.002

Béligon, V., Christophe, G., Fontanille, P., & Larroche, C. (2016). Microbial lipids as potential source to food supplements. *Current Opinion in Food Science*, *7*, 35–42. https://doi.org/10.1016/j.cofs.2015.10.002

Bahari, A., & Akoh, C. C. (2018a). Synthesis of a cocoa butter equivalent by enzymatic interesterification of illipe butter and palm mid-fraction. *Journal of the American Oil Chemists' Society*, *95*(5), 547–555. https://doi.org/10.1002/aocs.12083

Bahari, A., & Akoh, C. C. (2018b). Texture, rheology and fat bloom study of 'chocolates' made from cocoa butter equivalent synthesized from illipe butter and palm mid-fraction. *LWT-Food Science and Technology*, *97*, 349–354. https://doi.org/10.1016/j.lwt.2018.07.013

Bai, L., Lv, S., Xiang, W., Huan, S., Mcclements, D. J., & Rojas, O. J. (2019). Oil-in-water pickering emulsions via microfluidization with cellulose nanocrystals: 2. *In vitro* lipid digestion. *Food Hydrocolloids*, *96*, 709–716. https://doi.org/10.1016/j.foodhyd.2019.04.039

Bandarra, N. M., Lopes, P. A., Martins, S. V., Ferreira, J., Alfaia, C. M., Rolo, E. A., … Guil-Guerrero, J. L. (2016). Docosahexaenoic acid at the sn-2 position of structured triacylglycerols improved n-3 polyunsaturated fatty acid assimilation in tissues of hamsters. *Nutrition Research*, *36*(5), 452–463. https://doi.org/10.1016/j.nutres.2015.12.015

Barbut, S., & Marangoni, A. (2019). Organogels use in meat processing—Effects of fat/oil type and heating rate. *Meat science*, *149*, 9–13. https://doi.org/10.1016/j.meatsci.2018.11.003

Barbut, S., Marangoni, A. G., Thode, U., & Tiensa, B. E. (2019). Using canola oil organogels as fat replacement in liver pate. *Journal of Food Science*, *84*(9), 2646–2651. https://doi.org/10.1111/1750-3841.14753

Barbut, S., Wood, J., & Marangoni, A. (2016a). Potential use of organogels to replace animal fat in comminuted meat products. *Meat Science*, *122*, 155–162. https://doi.org/10.1016/j.meatsci.2016.08.003

Barbut, S., Wood, J., & Marangoni, A. (2016b). Quality effects of using organogels in breakfast sausage. *Meat Science*, *122*, 84–89. https://doi.org/10.1016/j.meatsci.2016.07.022

Barbut, S., Wood, J., & Marangoni, A. G. (2016c). Effects of organogel hardness and formulation on acceptance of frankfurters. *Journal of Food Science*, *81*(9), C2183–C2188. https://doi.org/10.1111/1750-3841.13409

Bassan, N., Rodrigues, R. H., Monti, R., Tecelão, C., Ferreira-Dias, S., & Paula, A. V. (2019a). Enzymatic modification of grape (*Vitis vinifera* L.) seed oil aimig to obtain dietary triacylglycerols in a batch reactor. *LWT-Food Science and Technology*, *99*, 600–606. https://doi.org/10.1016/j.lwt.2018.05.013

Bassan, N., Rodrigues, R. H., Monti, R., Tecelao, C., Ferreira-Dias, S., & Paula, A. V. (2019b). Enzymatic modification of grapeseed (*Vitis vinifera* L.) oil aiming to obtain dietary triacylglycerols in a batch reactor. *LWT-Food Science and Technology*, *99*, 600–606. https://doi.org/10.1016/j.lwt.2018.05.013

Berger, K., & Idris, N. A. (2005). Formulation of zero-*trans* acid shortenings and margarines and other food fats with products of the oil palm. *Journal of the American Oil Chemists' Society*, *82*(11), 775–782. https://doi.org/10.1007/s11746-005-1143-9

Bergfreund, J., Bertsch, P., Kuster, S., & Fischer, P. (2018). Effect of oil hydrophobicity on the adsorption and rheology of β-lactoglobulin at oil–water interfaces. *Langmuir*, *34*(16), 4929–4936. https://doi.org/10.1021/acs.langmuir.8b00458

Berton-Carabin, C. C., Sagis, L., & Schroën, K. (2018). Formation, structure, and functionality of interfacial layers in food emulsions. *Annual Review of Food Science and Technology*, *9*, 551–587. https://doi.org/10.1146/annurev-food-030117-012405

Berton-Carabin, C. C., Ropers, M. H., & Genot, C. (2014). Lipid oxidation in oil-in-water emulsions: Involvement of the interfacial layer. *Comprehensive Reviews in Food Science and Food Safety*, *13*(5), 945–977. https://doi.org/10.1111/1541-4337.12097

Binks, B., Davies, C., Fletcher, P., & Sharp, E. (2010). Non-aqueous foams in lubricating oil systems. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *360*(1–3), 198–204. https://doi.org/10.1016/j.colsurfa.2010.02.028

Binks, B. P., Garvey, E. J., & Vieira, J. (2016). Whipped oil stabilised by surfactant crystals. *Chemical Science*, *7*, 2621–2632. https://doi.org/10.1039/C6SC00046K

Binks, B. P., Johnston, S. K., Sekine, T., & Tyowua, A. T. (2015). Particles at oil–air surfaces: powdered oil, liquid oil marbles, and oil foam. *ACS Applied Materials & Interfaces*, *7*(26), 14328–14337. https://doi.org/10.1021/acsami.5b02890

Binks, B. P., & Marinopoulos, I. (2017). Ultra-stable self-foaming oils. *Food Research International*, *95*, 28–37. https://doi.org/10.1016/j.foodres.2017.02.020

Biswas, N., Cheow, Y. L., Tan, C. P., & Siow, L. F. (2018). Physicochemical properties of enzymatically produced palm-oil-based cocoa butter substitute (CBS) with cocoa butter mixture. *European Journal of Lipid Science and Technology*, *120*(3), 1700205. https://doi.org/10.1002/ejlt.201700205

Bornscheuer, U. T. (2018). Enzymes in lipid modification. *Annual Review of Food Science and Technology*, *9*, 85–103. https://doi.org/10.1146/annurev-food-030117-012336

Cai, Z., Wei, Y., Wu, M., Guo, Y., Xie, Y., Tao, R., … Zhang, H. (2019). Lipase immobilized on layer-by-layer polysaccharide-coated Fe_3O_4@ SiO_2 microspheres as a reusable biocatalyst for the production of structured lipids. *ACS Sustainable Chemistry & Engineering*, *7*(7), 6685–6695. https://doi.org/10.1021/acssuschemeng.8b05786

Celus, M., Salvia-Trujillo, L., Kyomugasho, C., Maes, I., Van Loey, A. M., Grauwet, T., & Hendrickx, M. E. (2018). Structurally modified pectin for targeted lipid antioxidant capacity in linseed/sunflower oil-in-water emulsions. *Food Chemistry*, *241*, 86–96. https://doi.org/10.1016/j.foodchem.2017.08.056

Chen, B., Mcclements, D. J., & Decker, E. A. (2014). Impact of diacylglycerol and monoacylglycerol on the physical and chemical properties of stripped soybean oil. *Food Chemistry*, *142*, 365–372. https://doi.org/10.1016/j.foodchem.2013.07.070

Chen, C., Van Damme, I., & Terentjev, E. (2009). Phase behavior of C18 monoglyceride in hydrophobic solutions. *Soft Matter*, *5*, 432–439. https://doi.org/10.1039/B813216J

Chen, F., Liang, L., Zhang, Z., Deng, Z., Decker, E. A., & Mcclements, D. J. (2017a). Inhibition of lipid oxidation in nanoemulsions and filled microgels fortified with omega-3 fatty acids using casein as a natural antioxidant. *Food Hydrocolloids*, *63*, 240–248. https://doi.org/10.1016/j.foodhyd.2016.09.001

Chen, Q., Li, Y., Fu, J., Ma, X., Teng, Y., & Wang, Y. (2017b). Production of diacylglycerol-enriched oils by enzymatic interesterification and molecular distillation using soybean oil and distilled saturated monoacylglycerol. *European Journal of Lipid Science and Technology*, *119*(6), 1600332. https://doi.org/10.1002/ejlt.201600332

Cheong, L. Z., Guo, Z., Lue, B. M., Miklos, R., Song, S., Panpipat, W., & Xu, X. (2012). Surface active lipids as encapsulation agents and delivery vehicles. In M. U. Ahmad (Ed.), *Lipids in nanotechnology* (pp. 15–51). Urbana, IL: AOCS Press. https://doi.org/10.1016/B978-0-9818936-7-9.50005-6

Cheong, L. Z., Zhang, H., Xu, Y., & Xu, X. (2009). Physical characterization of lard partial acylglycerols and their effects on melting and crystallization properties of blends with rapeseed oil. *Journal of Agricultural and Food Chemistry*, *57*(11), 5020–5027. https://doi.org/10.1021/jf900665h

Co, E. D., & Marangoni, A. G. (2012). Organogels: An alternative edible oil-structuring method. *Journal of the American Oil Chemists Society*, *89*(5), 749–780. https://doi.org/10.1007/s11746-012-2049-3

Corstens, M. N., Bertoncarabin, C. C., De Vries, R., Troost, F. J., Masclee, A. a. M., & Schroen, K. (2017). Food-grade microencapsulation systems that may induce satiety via delayed lipolysis: A review. *Critical Reviews in Food Science and Nutrition*, *57*(10), 2218–2244. https://doi.org/10.1080/10408398.2015.1057634

Craven, R. J., & Lencki, R. W. (2011). Crystallization and polymorphism of 1, 3-acyl-palmitoyl-*rac*-glycerols. *Journal of the American Oil Chemists' Society*, *88*(8), 1113–1123. https://doi.org/10.1007/s11746-011-1769-0

Czernichow, S., Thomas, D., & Bruckert, E. (2010). n-6 Fatty acids and cardiovascular health: A review of the evidence for dietary intake recommendations. *British Journal of Nutrition*, *104*(6), 788–796. https://doi.org/10.1017/S0007114510002096

Da Silva, R. C., Soares, F. a. S. D. M., Maruyama, J. M., Dagostinho, N. R., Silva, Y. A., Rodrigues Ract, J. N., & Gioielli, L. A. (2017). Crystallisation of monoacylglycerols and triacylglycerols at different proportions: Kinetics and structure. *International Journal of Food Properties*, *20*(sup1), S385–S398. https://doi.org/10.1080/10942912.2017.1297950

Da Silva, T. L. T., Domingues, M. a. F., Chiu, M. C., & Goncalves, L. a. G. (2017b). Templating effects of dipalmitin on soft palm midfraction crystals. *International Journal of Food Properties*, *20*(sup1), 935–947. https://doi.org/10.1080/10942912.2017.1318290

Daels, E., Foubert, I., & Goderis, B. (2017). The effect of adding a commercial phytosterol ester mixture on the phase behavior of palm oil. *Food Research International*, *100*, 841–849. https://doi.org/10.1016/j.foodres.2017.08.015

Dassanayake, L. S. K., Kodali, D. R., & Ueno, S. (2011). Formation of oleogels based on edible lipid materials. *Current Opinion in Colloid & Interface Science*, *16*(5), 432–439. https://doi.org/10.1016/j.cocis.2011.05.005

Davidovich-Pinhas, M. (2019). Oil structuring using polysaccharides. *Current Opinion in Food Science*, *27*, 29–35. https://doi.org/10.1016/j.cofs.2019.04.006

Davidovich-Pinhas, M., Barbut, S., & Marangoni, A. G. (2015). The gelation of oil using ethyl cellulose. *Carbohydrate Polymers*, *117*, 869–878. https://doi.org/10.1016/j.carbpol.2014.10.035

Davidovich-Pinhas, M., Barbut, S., & Marangoni, A. G. (2016). Development, characterization, and utilization of food-grade polymer oleogels. *Annual Review of Food Science and Technology*, *7*(7), 65–91. https://doi.org/10.1146/annurev-food-041715-033225

De Clercq, N., Kadivar, S., Van De Walle, D., De Pelsmaeker, S., Ghellynck, X., & Dewettinck, K. (2017). Functionality of cocoa butter equivalents in chocolate products. *European Food Research and Technology*, *243*(2), 309–321. https://doi.org/10.1007/s00217-016-2745-6

De Paula, A. V., Nunes, G. F., De Castro, H. F., & Dos Santos, J. C. (2018). Performance of packed bed reactor on the enzymatic interesterification of milk fat with soybean oil to yield structure lipids. *International Dairy Journal*, *86*, 1–8. https://doi.org/10.1016/j.idairyj.2018.06.014

De Vries, A., Gomez, Y. L., Jansen, B., Van Der Linden, E., & Scholten, E. (2017a). Controlling agglomeration of protein aggregates for structure formation in liquid oil: A sticky business. *Acs Applied Materials & Interfaces*, *9*(11), 10136–10147. https://doi.org/10.1021/acsami.7b00443

De Vries, A., Gomez, Y. L., Van Der Linden, E., & Scholten, E. (2017b). The effect of oil type on network formation by protein aggregates into oleogels. *Rsc Advances*, *7*(19), 11803–11812. https://doi.org/10.1039/c7ra00396j

De Vries, A., Jansen, D., Van Der Linden, E., & Scholten, E. (2018). Tuning the rheological properties of protein-based oleogels by water addition and heat treatment. *Food Hydrocolloids*, *79*, 100–109. https://doi.org/10.1016/j.foodhyd.2017.11.043

De Vries, A., Wesseling, A., Van Der Linden, E., & Scholten, E. (2017c). Protein oleogels from heat-set whey protein aggregates.

Journal of Colloid and Interface Science, *486*, 75–83. https://doi.org/10.1016/j.jcis.2016.09.043

Decker, E. A., Mcclements, D. J., Bourlieu-Lacanal, C., Durand, E., Figueroa-Espinoza, M. C., Lecomte, J., & Villeneuve, P. (2017). Hurdles in predicting antioxidant efficacy in oil-in-water emulsions. *Trends in Food Science and Technology*, *67*, 183–194. https://doi.org/10.1016/j.tifs.2017.07.001

Del Mar Muñío, M., Robles, A., Esteban, L., González, P. A., & Molina, E. (2009). Synthesis of structured lipids by two enzymatic steps: Ethanolysis of fish oils and esterification of 2-monoacylglycerols. *Process Biochemistry*, *44*(7), 723–730. https://doi.org/10.1016/j.procbio.2009.03.002

Deloid, G., Sohal, I. S., Lorente, L. R., Molina, R. M., Pyrgiotakis, G., Stevanovic, A., … Demokritou, P. (2018). Reducing intestinal digestion and absorption of fat using a nature-derived biopolymer: Interference of triglyceride hydrolysis by nanocellulose. *ACS Nano*, *12*(7), 6469–6479. https://doi.org/10.1021/acsnano.8b03074

Demirkesen, I., & Mert, B. (2019). Recent developments of oleogel utilizations in bakery products. *Critical Reviews in Food Science and Nutrition*, https://doi.org/10.1080/10408398.2019.1649243

Dian, N. L. H. M., Hamid, R. A., Kanagaratnam, S., Isa, W. R. A., Hassim, N. a. M., Ismail, N. H., … Sahri, M. M. (2017). Palm oil and palm kernel oil: Versatile ingredients for food applications. *Journal of Oil Palm Research*, *29*(4), 487–511. https://doi.org/10.21894/jopr.2017.00014

Diao, X., Guan, H., Zhao, X., Chen, Q., & Kong, B. (2016). Properties and oxidative stability of emulsions prepared with myofibrillar protein and lard diacylglycerols. *Meat Science*, *115*, 16–23. https://doi.org/10.1016/j.meatsci.2016.01.001

Dicosimo, R., Mcauliffe, J., Poulose, A. J., & Bohlmann, G. (2013). Industrial use of immobilized enzymes. *Chemical Society Reviews*, *42*(15), 6437–6474. https://doi.org/10.1039/C3CS35506C

Doan, C. D., Tavernier, I., Okuro, P. K., & Dewettinck, K. (2018). Internal and external factors affecting the crystallization, gelation and applicability of wax-based oleogels in food industry. *Innovative Food Science & Emerging Technologies*, *45*, 42–52. https://doi.org/10.1016/j.ifset.2017.09.023

Domingues, M. a. F., Da Silva, T. L. T., Ribeiro, A. P. B., Chiu, M. C., & Gonçalves, L. a. G. (2016). Sucrose behenate as a crystallization enhancer for soft fats. *Food Chemistry*, *192*, 972–978. https://doi.org/10.1016/j.foodchem.2015.07.109

Dong, Z., Liu, Z., Shi, J., Tang, H., Xiang, X., Huang, F., & Zheng, M. (2019). Carbon nanoparticle-stabilized pickering emulsion as a sustainable and high-performance interfacial catalysis platform for enzymatic esterification/transesterification. *ACS Sustainable Chemistry & Engineering*, *7*(8), 7619–7629. https://doi.org/10.1021/acssuschemeng.8b05908

Facin, B. R., Melchiors, M. S., ValéRio, A., Oliveira, J. V., & Oliveira, D. B. D. (2019). Driving immobilized lipases as biocatalysts: 10 years state of the art and future prospects. *Industrial & Engineering Chemistry Research*, *58*(14), 5358–5378. https://doi.org/10.1021/acs.iecr.9b00448

Fameau, A. L., Lam, S., Arnould, A., Gaillard, C. D., Velev, O. D., & Saint-Jalmes, A. (2015). Smart nonaqueous foams from lipid-based oleogel. *Langmuir*, *31*(50), 13501–13510. https://doi.org/10.1021/acs.langmuir.5b03660

Fameau, A. L., & Saint-Jalmes, A. (2017). Non-aqueous foams: Current understanding on the formation and stability mechanisms. *Advances in Colloid and Interface Science*, *247*, 454–464. https://doi.org/10.1016/j.cis.2017.02.007

Faustino, A. R., Osório, N. M., Tecelão, C., Canet, A., Valero, F., & Ferreira-Dias, S. (2016). Camelina oil as a source of polyunsaturated fatty acids for the production of human milk fat substitutes catalyzed by a heterologous *Rhizopus oryzae* lipase. *European Journal of Lipid Science and Technology*, *118*(4), 532–544. https://doi.org/10.1002/ejlt.201500003

Feltes, M. M. C., De Oliveira, D., Block, J. M., & Ninow, J. L. (2013). The production, benefits, and applications of monoacylglycerols and diacylglycerols of nutritional interest. *Food and Bioprocess Technology*, *6*(1), 17–35. https://doi.org/10.1007/s11947-012-0836-3

Ferreira, M. L., & Tonetto, G. M. (2017a). *Enzymatic synthesis of structured triglycerides: From laboratory to industry*. Cham, Switzerland: Springer. https://doi.org/10.1007/978-3-319-51574-8

Ferreira, M. L., & Tonetto, G. M. (2017b). What is the importance of structured triglycerides and diglycerides? *Enzymatic synthesis of structured triglycerides* (pp. 1–16). Cham, Switzerland: Springer. https://doi.org/10.1007/978-3-319-51574-8_1

Flickinger, B. D., & Matsuo, N. (2005). Diacylglycerols. In F. Shahidi (Ed.), *Bailey's industrial oil and fat products*. Hoboken, NJ: Wiley. https://doi.org/10.1002/047167849X.bio069

Freiría-Gándara, J., Losada-Barreiro, S., Paiva-Martins, F., & Bravo-Díaz, C. (2018). Enhancement of the antioxidant efficiency of gallic acid derivatives in intact fish oil-in-water emulsions through optimization of their interfacial concentrations. *Food & Function*, *9*(8), 4429–4442. https://doi.org/10.1039/C8FO00977E

Ganesan, B., Brothersen, C., & Mcmahon, D. J. (2014). Fortification of foods with omega-3 polyunsaturated fatty acids. *Critical Reviews in Food Science and Nutrition*, *54*(1), 98–114. https://doi.org/10.1080/10408398.2011.578221

Gao, Z. M., Yang, X. Q., Wu, N. N., Wang, L. J., Wang, J. M., Guo, J., & Yin, S. W. (2014). Protein-based pickering emulsion and oil gel prepared by complexes of zein colloidal particles and stearate. *Journal of Agricultural and Food Chemistry 62*(12), 2672–2678. https://doi.org/10.1021/jf500005y

Ghosh, M., Sengupta, A., Bhattacharyya, D., & Ghosh, M. (2016). Preparation of human milk fat analogue by enzymatic interesterification reaction using palm stearin and fish oil. *Journal of Food Science and Technology-Mysore*, *53*(4), 2017–2024. https://doi.org/10.1007/s13197-016-2180-5

Giacintucci, V., Mattia, C., Sacchetti, G., Flamminii, F., Gravelle, A., Baylis, B., … Pittia, P. (2018). Ethylcellulose oleogels with extra virgin olive oil: the role of oil minor components on microstructure and mechanical strength. *Food Hydrocolloids*, *84*, 508–514. https://doi.org/10.1016/j.foodhyd.2018.05.030

Gollaher, C. J., & Bistrian, B. R. (2018). Structured lipids. In A. R. Forse (Ed.), *Diet Nutrition and Immunity* (pp. 187–198). Boca Raton, FL: CRC Press. https://doi.org/10.1201/9781351071390

Gomez-Estaca, J., Herrero, A. M., Herranz, B., Alvarez, M. D., Jimenez-Colmenero, F., & Cofrades, S. (2019a). Characterization of ethyl cellulose and beeswax oleogels and their suitability as fat replacers in healthier lipid pates development. *Food Hydrocolloids*, *87*, 960–969. https://doi.org/10.1016/j.foodhyd.2018.09.029

Gomez-Estaca, J., Pintado, T., Jimenez-Colmenero, F., & Cofrades, S. (2019b). Assessment of a healthy oil combination structured in ethyl cellulose and beeswax oleogels as animal fat replacers in low-fat, PUFA-enriched pork burgers. *Food and Bioprocess Technology*, *12*(6), 1068–1081. https://doi.org/10.1007/s11947-019-02281-3

Gonzalez-Gutierrez, J., & Scanlon, M. G. (2018). Rheology and mechanical properties of fats. In A. G. Marangoni (Ed.), *Structure-Function*

Analysis of Edible Fats (pp. 119–168). Urbana, IL: AOCS Press. https://doi.org/10.1016/B978-0-12-814041-3.00005-8

Gravelle, A. J., Blach, C., Weiss, J., Barbut, S., & Marangoni, A. G. (2017). Structure and properties of an ethylcellulose and stearyl alcohol/stearic acid (EC/SO:SA) hybrid oleogelator system. *European Journal of Lipid Science and Technology*, *119*(11), 1700069. Retrieved from https://doi.org/Artn170006910.1002/Ejlt.201700069

Gravelle, A. J., Davidovich-Pinhas, M., Barbut, S., & Marangoni, A. G. (2017). Influencing the crystallization behavior of binary mixtures of stearyl alcohol and stearic acid (SOSA) using ethylcellulose. *Food Research International*, *91*, 1–10. https://doi.org/10.1016/j.foodres.2016.11.024

Gravelle, A. J., Davidovich-Pinhas, M., Zetzl, A. K., Barbut, S., & Marangoni, A. G. (2016). Influence of solvent quality on the mechanical strength of ethylcellulose oleogels. *Carbohydrate Polymers*, *135*, 169–179. https://doi.org/10.1016/j.carbpol.2015.08.050

Gravelle, A. J., & Marangoni, A. G. (2018). Ethylcellulose oleogels: Structure, functionality, and food applications. *Advances in Food and Nutrition Research 84*, 1–56. https://doi.org/10.1016/bs.afnr.2018.01.002

Gumus, C. E., Decker, E. A., & Mcclements, D. J. (2017). Impact of legume protein type and location on lipid oxidation in fish oil-in-water emulsions: Lentil, pea, and faba bean proteins. *Food Research International*, *100*, 175–185. https://doi.org/10.1016/j.foodres.2017.08.029

Gunes, D., Murith, M., Godefroid, J., Pelloux, C., Deyber, H., Schafer, O., & Breton, O. (2017). Oleofoams: Properties of crystal-coated bubbles from whipped oleogels-evidence for pickering stabilization. *Langmuir*, *33*(6), 1563–1575. https://doi.org/10.1021/acs.langmuir.6b04141

Guo, Q., Ye, A., Bellissimo, N., Singh, H., & Rousseau, D. (2017). Modulating fat digestion through food structure design. *Progress in Lipid Research*, *68*, 109–118. https://doi.org/10.1016/j.plipres.2017.10.001

Gupta, A., Barrow, C. J., & Puri, M. (2012). Omega-3 biotechnology: Thraustochytrids as a novel source of omega-3 oils. *Biotechnology Advances*, *30*(6), 1733–1745. https://doi.org/10.1016/j.biotechadv.2012.02.014

Hagemann, J. (1988). Thermal behavior and polymorphism of acylglycerides. In N. Garti & K. Sato (Eds.), *Crystallization and polymorphism of fat and fatty acids* (pp. 9–95). New York, NY: Marcel Dekker Inc.

Hartel, R. W. (2013). Advances in food crystallization. *Annual Review of Food Science and Technology*, *4*, 277–292. https://doi.org/10.1146/annurev-food-030212-182530

He, Y., Li, J., Guo, Z., & Chen, B. (2018). Synthesis of novel medium-long-medium type structured lipids from microalgae oil via two-step enzymatic reactions. *Process Biochemistry*, *68*, 108–116. https://doi.org/10.1016/j.procbio.2018.02.005

He, Y., Li, J., Kodali, S., Balle, T., Chen, B., & Guo, Z. (2017a). Liquid lipases for enzymatic concentration of n-3 polyunsaturated fatty acids in monoacylglycerols via ethanolysis: Catalytic specificity and parameterization. *Bioresource Technology*, *224*, 445–456. https://doi.org/10.1016/j.biortech.2016.10.087

He, Y., Qiu, C., Guo, Z., Huang, J., Wang, M., & Chen, B. (2017b). Production of new human milk fat substitutes by enzymatic acidolysis of microalgae oils from Nannochloropsis oculata and Isochrysis galbana. *Bioresource Technology*, *238*, 129–138. https://doi.org/10.1016/j.biortech.2017.04.041

He, Y. J., Qiu, C. Y., Guo, Z., Huang, J., Wang, M. Z., & Chen, B. L. (2017c). Production of new human milk fat substitutes by enzymatic acidolysis of microalgae oils from *Nannochloropsis oculata* and *Isochrysis galbana*. *Bioresource Technology*, *238*, 129–138. https://doi.org/10.1016/j.biortech.2017.04.041

Hermund, D. B., Karadagˇ, A. E., Andersen, U., Jónsdóttir, R., Kristinsson, H. G., Alasalvar, C., & Jacobsen, C. (2016). Oxidative stability of granola bars enriched with multilayered fish oil emulsion in the presence of novel brown seaweed based antioxidants. *Journal of Agricultural and Food Chemistry*, *64*(44), 8359–8368. https://doi.org/10.1021/acs.jafc.6b03454

Heymans, R., Tavernier, I., Danthine, S., Rimaux, T., Van Der Meeren, P., & Dewettinck, K. (2018). Food-grade monoglyceride oil foams: The effect of tempering on foamability, foam stability and rheological properties. *Food & Function*, *9*(6), 3143–3154. https://doi.org/10.1039/C8FO00536B

Heymans, R., Tavernier, I., Dewettinck, K., & Meeren, P. V. D. (2017). Crystal stabilization of edible oil foams. *Trends in Food Science and Technology*, *69*, 13–24. https://doi.org/10.1016/j.tifs.2017.08.015

Hu, P., Xu, X., & Yu, L. L. (2017). Interesterified *trans*-free fats rich in sn-2 nervonic acid prepared using Acer truncatum oil, palm stearin and palm kernel oil, and their physicochemical properties. *LWT-Food Science and Technology*, *76*, 156–163. https://doi.org/10.1016/j.lwt.2016.10.054

Huang, Y., He, M., Lu, A., Zhou, W. Z., Stoyanov, S. D., Pelan, E. G., & Zhang, L. N. (2015). Hydrophobic modification of chitin whisker and its potential application in structuring oil. *Langmuir*, *31*(5), 1641–1648. https://doi.org/10.1021/la504576p

Ilyasoglu, H. (2013). Production of human fat milk analogue containing α-linolenic acid by solvent-free enzymatic interesterification. *LWT-Food Science and Technology*, *54*(1), 179–185. https://doi.org/10.1016/j.lwt.2013.05.036

Jahurul, M., Zaidul, I., Norulaini, N., Sahena, F., Jinap, S., Azmir, J., … Omar, A. M. (2013). Cocoa butter fats and possibilities of substitution in food products concerning cocoa varieties, alternative sources, extraction methods, composition, and characteristics. *Journal of Food Engineering*, *117*(4), 467–476. https://doi.org/10.1016/j.jfoodeng.2012.09.024

Jiang, Y., Liu, L. L., Wang, B. J., Sui, X. F., Zhong, Y., Zhang, L. P., … Xu, H. (2018). Cellulose-rich oleogels prepared with an emulsion-templated approach. *Food Hydrocolloids*, *77*, 460–464. https://doi.org/10.1016/j.foodhyd.2017.10.023

Jimenez-Colmenero, F., Salcedo-Sandoval, L., Bou, R., Cofrades, S., Herrero, A. M., & Ruiz-Capillas, C. (2015). Novel applications of oil-structuring methods as a strategy to improve the fat content of meat products. *Trends in Food Science & Technology*, *44*(2), 177–188. https://doi.org/10.1016/j.tifs.2015.04.011

Jin, J., Jin, Q. Z., Wang, X. G., & Akoh, C. C. (2019). Improving heat and fat bloom stabilities of "dark chocolates" by addition of mango kernel fat-based chocolate fats. *Journal of Food Engineering*, *246*, 33–41. https://doi.org/10.1016/j.jfoodeng.2018.10.027

Johnson, D. R., Inchingolo, R., & Decker, E. A. (2018). The ability of oxygen scavenging packaging to inhibit vitamin degradation and lipid oxidation in fish oil-in-water emulsions. *Innovative Food Science and Emerging Technologies*, *47*, 467–475. https://doi.org/10.1016/j.ifset.2018.04.021

Kabri, T. H., Meynier, A., Bontemps, D., Gaillard, C., Foucat, L., Linder, M., & Genot, C. (2013). Formulation of sub-micron emulsions containing docosahexaenoic acid esterified in triacylglycerols or

phospholipids. *European Journal of Lipid Science and Technology*, *115*(11), 1294–1308. https://doi.org/doi.org/10.1002/ejlt.201300045

Kadamne, J. V., Ifeduba, E. A., Akoh, C. C., & Martini, S. (2017). Sonocrystallization of interesterified fats with 20 and 30% C16: 0 at *sn*-2 position. *Journal of the American Oil Chemists' Society*, *94*(1), 3–18. https://doi.org/10.1007/s11746-016-2914-6

Kadhum, 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

Kim, B. H., & Akoh, C. C. (2006). Characteristics of structured lipid prepared by lipase-catalyzed acidolysis of roasted sesame oil and caprylic acid in a bench-scale continuous packed bed reactor. *Journal of Agricultural and Food Chemistry*, *54*(14), 5132–5141. https://doi.org/10.1021/jf060607k

Kim, B. H., & Akoh, C. C. (2015). Recent research trends on the enzymatic synthesis of structured lipids. *Journal of Food Science*, *80*(8), C1713–C1724. https://doi.org/10.1111/1750-3841.12953

Kristensen, J. B., Nielsen, N. S., Jacobsen, C., & Mu, H. (2006). Oxidative stability of diacylglycerol oil and butter blends containing diacylglycerols. *European Journal of Lipid Science and Technology*, *108*(4), 336–350. https://doi.org/10.1002/ejlt.200500232

Krog, N. (2001). Crystallization properties and lyotropic phase behavior of food emulsifiers: Relation to technical applications. In N. Garti & K. Sato (Eds.), *Crystallization processes in fats and lipid systems* (pp. 519–540). Boca Raton, FL: CRC Press. https://doi.org/10.1201/9781482270884

Krog, N. J., & Sparsø, F. V. (2003). Food emulsifiers: Their chemical and physical properties. In S. E. Friberg, K. Larsson, & J. Sjöblom (Eds.), *Food emulsions* (pp. 60–106). New York, NY: Marcel Dekker Inc. https://doi.org/10.1201/9780203913222

Lakum, R., & Sonwai, S. (2018). Production of *trans*-free margarine fat by enzymatic interesterification of soy bean oil, palm stearin and coconut stearin blend. *International Journal of Food Science and Technology*, *53*(12), 2761–2769. https://doi.org/10.1111/ijfs.13888

Lee, J., & Decker, E. A. (2011). Effects of metal chelator, sodium azide, and superoxide dismutase on the oxidative stability in riboflavin-photosensitized oil-in-water emulsion systems. *Journal of Agricultural and Food Chemistry*, *59*(11), 6271–6276. https://doi.org/10.1021/jf2001537

Lee, K. T., & Akoh, C. C. (1998). Structured lipids: Synthesis and applications. *Food Reviews International*, *14*(1), 17–34. https://doi.org/https://doi.org/10.1080/87559129809541148

Lee, Y. Y., Tang, T. K., Phuah, E. T., Tan, C. P., Wang, Y., Li, Y., … Lai, O. M. (2019). Production, safety, health effects and applications of diacylglycerol functional oil in food systems: A review. *Critical Reviews in Food Science and Nutrition*, https://doi.org/10.1080/10408398.2019.1650001

Lesmes, U., Sandra, S., Decker, E. A., & Mcclements, D. J. (2010). Impact of surface deposition of lactoferrin on physical and chemical stability of omega-3 rich lipid droplets stabilised by caseinate. *Food Chemistry*, *123*(1), 99–106. https://doi.org/10.1016/j.foodchem.2010.04.007

Li, D., Qin, X., Wang, J., Yang, B., Wang, W., Huang, W., & Wang, Y. (2015a). Hydrolysis of soybean oil to produce diacylglycerol by a lipase from *Rhizopus oryzae*. *Journal of Molecular Catalysis B: Enzymatic*, *115*, 43–50. https://doi.org/10.1016/j.molcatb.2015.01.009

Li, G., Chen, J., Ma, X., Zhang, Z., Liu, N., & Wang, Y. (2018a). Enzymatic preparation and facile purification of medium-chain, and medium-and long-chain fatty acid diacylglycerols. *LWT-Food Science and Technology*, *92*, 227–233. https://doi.org/10.1016/j.lwt.2018.02.032

Li, R. Y., Sabir, J. S. M., Baeshen, N. A., & Akoh, C. C. (2015b). Enzymatic synthesis of refined olive oil-based structured lipid containing omega-3 and-6 fatty acids for potential application in infant formula. *Journal of Food Science*, *80*(11), H2578–H2584. https://doi.org/10.1111/1750-3841.13075

Li, Y., Zhao, J., Xie, X., Zhang, Z., Zhang, N., & Wang, Y. (2018b). 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*, 405–413. https://doi.org/10.1016/j.foodchem.2018.02.086

Lian, W., Wang, W., Tan, C. P., Wang, J., & Wang, Y. (2019). Immobilized *Talaromyces thermophilus* lipase as an efficient catalyst for the production of LML-type structured lipids. *Bioprocess and Biosystems Engineering*, *42*(2), 321–329. https://doi.org/10.1007/s00449-018-2036-7

Lin, Z., & Goddard, J. (2018). Photo-curable metal-chelating coatings offer a scalable approach to production of antioxidant active packaging. *Journal of Food Science*, *83*(2), 367–376. https://doi.org/10.1111/1750-3841.14051

Litwinenko, J. W., Rojas, A. M., Gerschenson, L. N., & Marangoni, A. G. (2002). Relationship between crystallization behavior, microstructure, and mechanical properties in a palm oil-based shortening. *Journal of the American Oil Chemists Society*, *79*(7), 647–654. https://doi.org/10.1007/s11746-002-0538-y

Liu, L., Kerr, W. L., & Kong, F. (2019). Characterization of lipid emulsions during *in vitro* digestion in the presence of three types of nanocellulose. *Journal of Colloid and Interface Science*, *545*, 317–329. https://doi.org/10.1016/j.jcis.2019.03.023

Liu, M., Fu, J., Teng, Y., Zhang, Z., Zhang, N., & Wang, Y. (2016). Fast production of diacylglycerol in a solvent free system via lipase catalyzed esterification using a bubble column reactor. *Journal of the American Oil Chemists' Society*, *93*(5), 637–648. https://doi.org/10.1007/s11746-016-2804-y

Liu, N., Li, D., Wang, W., Hollmann, F., Xu, L., Ma, Y., … Wang, Y. (2018). Production and immobilization of lipase PCL and its application in synthesis of α-linolenic acid-rich diacylglycerol. *Journal of Food Biochemistry*, *42*(5), e12574. https://doi.org/10.1111/jfbc.12574

Lo, S. K., Tan, C. P., Long, K., Yusoff, M. S. A., & Lai, O. M. (2008). Diacylglycerol oil—Properties, processes and products: A review. *Food and Bioprocess Technology*, *1*(3), 223–233. https://doi.org/10.1007/s11947-007-0049-3

Long, Z., Zhao, M., Liu, N., Liu, D., Sun-Waterhouse, D., & Zhao, Q. (2015). Physicochemical properties of peanut oil-based diacylglycerol and their derived oil-in-water emulsions stabilized by sodium caseinate. *Food Chemistry*, *184*, 105–113. https://doi.org/10.1016/j.foodchem.2015.03.052

Luo, S. Z., Hu, X. F., Jia, Y. J., Pan, L. H., Zheng, Z., Zhao, Y. Y., … Jiang, S. T. (2019). Camellia oil-based oleogels structuring with tea polyphenol-palmitate particles and citrus pectin by emulsion-templated method: Preparation, characterization and potential application. *Food Hydrocolloids*, *95*, 76–87. https://doi.org/10.1016/j.foodhyd.2019.04.016

Ma, X., Jiang, Z., & Lai, C. (2016). Significance of increasing n-3 PUFA content in pork on human health. *Critical Reviews in Food Science and Nutrition*, *56*(5), 858–870. https://doi.org/10.1080/10408398.2013.850059

Macias-Rodriguez, B. A., & Marangoni, A. A. (2018). Linear and nonlinear rheological behavior of fat crystal networks. *Critical Reviews in Food Science and Nutrition*, *58*(14), 2398–2415. https://doi.org/10.1080/10408398.2017.1325835

Makeri, M., Sahri, M. M., Ghazali, H. M., Ahmad, K., & Muhammad, K. (2019). Polymorphism, textural and crystallization properties of winged bean (*Psophocarpus tetragonolobus*, D.C) oil-based *trans*-fatty acids free ternary margarine blends. *LWT-Food Science and Technology*, *100*, 158–166. https://doi.org/10.1016/j.lwt.2018.09.012

Marangoni, A. G., Acevedo, N., Maleky, F., Co, E., Peyronel, F., Mazzanti, G., … Pink, D. (2012). Structure and functionality of edible fats. *Soft Matter*, *8*(5), 1275–1300. https://doi.org/10.1039/C1SM06234D

Martín, M. P., Grosso, A. L., Nepote, V., & Grosso, N. R. (2018). Sensory and chemical stabilities of high-oleic and normal-oleic peanuts in shell during long-term storage. *Journal of Food Science*, *83*(9), 2362–2368. https://doi.org/10.1111/1750-3841.14295

Martins, A. J., Vicente, A. A., Cunha, R. L., & Cerqueira, M. A. (2018). Edible oleogels: An opportunity for fat replacement in foods. *Food & Function*, *9*(2), 758–773. https://doi.org/10.1039/c7fo01641g

Mateo, C., Palomo, J. M., Fernandez-Lorente, G., Guisan, J. M., & Fernandez-Lafuente, R. (2007). Improvement of enzyme activity, stability and selectivity via immobilization techniques. *Enzyme and Microbial Technology*, *40*(6), 1451–1463. https://doi.org/10.1016/j.enzmictec.2007.01.018

Matsuo, N. (2007). Nutritional characteristics and health benefits of diacylglycerol in foods. *Food Science and Technology Research*, *10*(2), 103–110. https://doi.org/10.3136/fstr.10.103

Mcclements, D. J. (2018). The biophysics of digestion: Lipids. *Current Opinion in Food Science*, *21*, 1–6. https://doi.org/10.1016/j.cofs.2018.03.009

Mcclements, D. J., & Decker, E. (2017). Interfacial antioxidants: A review of natural and synthetic emulsifiers and coemulsifiers that can inhibit lipid oxidation. *Journal of Agricultural and Food Chemistry*, *66*(1), 20–35. https://doi.org/10.1021/acs.jafc.7b05066

Meng, Z., Qi, K. Y., Guo, Y., Wang, Y., & Liu, Y. F. (2018a). Effects of thickening agents on the formation and properties of edible oleogels based on hydroxypropyl methyl cellulose. *Food Chemistry*, *246*, 137–149. https://doi.org/10.1016/j.foodchem.2017.10.154

Meng, Z., Qi, K. Y., Guo, Y., Wang, Y., & Liu, Y. F. (2018b). Macro-micro structure characterization and molecular properties of emulsion-templated polysaccharide oleogels. *Food Hydrocolloids*, *77*, 17–29. https://doi.org/10.1016/j.foodhyd.2017.09.006

Meng, Z., Qi, K. Y., Guo, Y., Wang, Y., & Liu, Y. F. (2018c). Physical properties, microstructure, intermolecular forces, and oxidation stability of soybean oil oleogels structured by different cellulose ethers. *European Journal of Lipid Science and Technology*, *120*(6), 1700287. https://doi.org/Artn170028710.1002/Ejlt.201700287

Merrill, L. I., Pike, O. A., Ogden, L. V., & Dunn, M. L. (2008). Oxidative stability of conventional and high-oleic vegetable oils with added antioxidants. *Journal of the American Oil Chemists' Society*, *85*(8), 771–776. https://doi.org/10.1007/s11746-008-1256-4

Metin, S., & Hartel, R. W. (1998). Thermal analysis of isothermal crystallization kinetics in blends of cocoa butter with milk fat or milk fat fractions. *Journal of the American Oil Chemists Society*, *75*(11), 1617–1624. https://doi.org/10.1007/s11746-998-0102-z

Metin, S., & Hartel, R. W. (2005). Crystallization of fats and oils. In F. Shahidi (Ed.), *Bailey's industrial oil and fat products*. Hoboken, NJ: John Wiley & Sons. https://doi.org/10.1002/047167849X.bio021

Miklos, R., Zhang, H., Lametsch, R., & Xu, X. (2013). Physicochemical properties of lard-based diacylglycerols in blends with lard. *Food Chemistry*, *138*(1), 608–614. https://doi.org/10.1016/j.foodchem.2012.10.070

Mishima, S., Suzuki, A., Sato, K., & Ueno, S. (2016). Formation and microstructures of whipped oils composed of vegetable oils and high-melting fat crystals. *Journal of the American Oil Chemists' Society*, *93*(11), 1453–1466. https://doi.org/10.1007/s11746-016-2888-4

Monteiro, J., Leslie, M., Moghadasian, M. H., Arendt, B. M., Allard, J. P., & Ma, D. W. (2014). The role of n−6 and n−3 polyunsaturated fatty acids in the manifestation of the metabolic syndrome in cardiovascular disease and non-alcoholic fatty liver disease. *Food & Function*, *5*(3), 426–435. https://doi.org/10.1039/C3FO60551E

Munk, M. B., Utoft, A., Larsen, F. H., Needham, D., & Risbo, J. (2019). Oleogelating properties of ethylcellulose in oil-in-water emulsions: The impact of emulsification methods studied by C-13 MAS NMR, surface tension and micropipette manipulation studies. *Food Hydrocolloids*, *89*, 700–706. https://doi.org/10.1016/j.foodhyd.2018.11.019

Nagavekar, N., Kumar, A., Dubey, K., & Singhal, R. S. (2019). Supercritical carbon dioxide extraction of kokum fat from Garcinia indica kernels and its application as a gelator in oleogels with oils. *Industrial Crops and Products*, *138*, 111459. https://doi.org/UNSP11145910.1016/j.indcrop.2019.06.022

Napolitano, G. E., Ye, Y., & Cruz-Hernandez, C. (2018). Chemical characterization of a high-oleic soybean oil. *Journal of the American Oil Chemists' Society*, *95*(5), 583–589. https://doi.org/10.1002/aocs.12049

Nepote, V., Olmedo, R. H., Mestrallet, M. G., & Grosso, N. (2009). A study of the relationships among consumer acceptance, oxidation chemical indicators, and sensory attributes in high-oleic and normal peanuts. *Journal of Food Science*, *74*(1), S1–S8. https://doi.org/10.1111/j.1750-3841.2008.00972.x

Ng, S. P., Lai, O. M., Abas, F., Lim, H. K., & Tan, C. P. (2014). Stability of a concentrated oil-in-water emulsion model prepared using palm olein-based diacylglycerol/virgin coconut oil blends: Effects of the rheological properties, droplet size distribution and microstructure. *Food Research International*, *64*, 919–930. https://doi.org/10.1016/j.foodres.2014.08.045

Nicholson, T., Khademi, H., & Moghadasian, M. H. (2013). The role of marine n-3 fatty acids in improving cardiovascular health: A review. *Food & Function*, *4*(3), 357–365. https://doi.org/10.1039/C2FO30235G

Nielsen, N. S., Petersen, A., Meyer, A. S., Timm-Heinrich, M., & Jacobsen, C. (2004). Effects of lactoferrin, phytic acid, and EDTA on oxidation in two food emulsions enriched with long-chain polyunsaturated fatty acids. *Journal of Agricultural and Food Chemistry*, *52*(25), 7690–7699. https://doi.org/10.1021/jf0492316

Nikiforidis, C. V., & Scholten, E. (2015). Polymer organogelation with chitin and chitin nanocrystals. *RSC Advances*, *5*(47), 37789–37799. https://doi.org/10.1039/c5ra06451a

Norizzah, A., Chong, C., Cheow, C., & Zaliha, O. (2004). Effects of chemical interesterification on physicochemical properties of palm

stearin and palm kernel olein blends. *Food Chemistry*, *86*(2), 229–235. https://doi.org/10.1016/j.foodchem.2003.09.030

Norn, V. (2015). *Emulsifiers in food technology*. Hoboken, NJ: John Wiley & Sons.

Nunes, P. A., Pires-Cabral, P., & Ferreira-Dias, S. (2011). Production of olive oil enriched with medium chain fatty acids catalysed by commercial immobilised lipases. *Food Chemistry*, *127*(3), 993–998. https://doi.org/10.1016/j.foodchem.2011.01.071

O'sullivan, C. M., Barbut, S., & Marangoni, A. G. (2016). Edible oleogels for the oral delivery of lipid soluble molecules: Composition and structural design considerations. *Trends in Food Science & Technology*, *57*, 59–73. https://doi.org/10.1016/j.tifs.2016.08.018

O'sullivan, C. M., Davidovich-Pinhas, M., Wright, A. J., Barbut, S., & Marangoni, A. G. (2017). Ethylcellulose oleogels for lipophilic bioactive delivery—Effect of oleogelation on in vitro bioaccessibility and stability of beta-carotene. *Food & Function*, *8*(4), 1438–1451. https://doi.org/10.1039/c6fo01805j

Oh, I., Lee, J., Lee, H. G., & Lee, S. (2019). Feasibility of hydroxypropyl methylcellulose oleogel as an animal fat replacer for meat patties. *Food Research International*, *122*, 566–572. https://doi.org/10.1016/j.foodres.2019.01.012

Oh, I. K., & Lee, S. (2018). Utilization of foam structured hydroxypropyl methylcellulose for oleogels and their application as a solid fat replacer in muffins. *Food Hydrocolloids*, *77*, 796–802. https://doi.org/10.1016/j.foodhyd.2017.11.022

Ornla-Ied, P., Sonwai, S., & Lertthirasuntorn, S. (2016). Trans-free margarine fat produced using enzymatic interesterification of rice bran oil and hard palm stearin. *Food Science and Biotechnology*, *25*(3), 673–680. https://doi.org/10.1007/s10068-016-0118-3

Osborn, H., & Akoh, C. (2002). Structured lipids-novel fats with medical, nutraceutical, and food applications. *Comprehensive Reviews in Food Science and Food Safety*, *1*(3), 110–120. https://doi.org/10.1111/j.1541-4337.2002.tb00010.x

Palacios, D., Ortega, N., Rubio-Rodríguez, N., & Busto, M. D. (2019). Lipase-catalyzed glycerolysis of anchovy oil in a solvent-free system: Simultaneous optimization of monoacylglycerol synthesis and end-product oxidative stability. *Food Chemistry*, *271*, 372–379. https://doi.org/10.1016/j.foodchem.2018.07.184

Pande, G., & Akoh, C. C. (2013). Enzymatic modification of lipids for trans-free margarine. *Lipid Technology*, *25*(2), 31–33. https://doi.org/10.1002/lite.201300253

Pande, G., Akoh, C. C., & Shewfelt, R. L. (2012). Production of *trans*-free margarine with stearidonic acid soybean and high-stearate soybean oils-based structured lipid. *Journal of Food Science*, *77*(11), C1203–C1210. https://doi.org/10.1111/j.1750-3841.2012.02935.x

Patel, A. R. (2017a). A colloidal gel perspective for understanding oleogelation. *Current Opinion in Food Science*, *15*, 1–7. https://doi.org/10.1016/j.cofs.2017.02.013

Patel, A. R. (2017b). Edible "oleocolloids": The final frontier in food innovation? *Journal of Agricultural and Food Chemistry*, *65*(17), 3432–3433. https://doi.org/10.1021/acs.jafc.7b01421

Patel, A. R. (2017c). Methylcellulose-coated microcapsules of Palm stearine as structuring templates for creating hybrid oleogels. *Materials Chemistry and Physics*, *195*, 268–274. https://doi.org/10.1016/j.matchemphys.2017.03.059

Patel, A. R. (2017d). Stable 'arrested' non-aqueous edible foams based on food emulsifiers. *Food & Function*, *8*(6), 2115–2120. https://doi.org/10.1039/c7fo00187h

Patel, A. R. (2018). Structuring edible oils with hydrocolloids: Where do we stand? *Food Biophysics*, *13*(2), 113–115. https://doi.org/10.1007/s11483-018-9527-6

Patel, A. R., Cludts, N., Bin Sintang, M. D., Lesaffer, A., & Dewettinck, K. (2014a). Edible oleogels based on water soluble food polymers: Preparation, characterization and potential application. *Food & Function*, *5*(11), 2833–2841. https://doi.org/10.1039/c4fo00624k

Patel, A. R., Cludts, N., Bin Sintang, M. D., Lewille, B., Lesaffer, A., & Dewettinck, K. (2014b). Polysaccharide-based oleogels prepared with an emulsion-templated approach. *Chemphyschem*, *15*(16), 3435–3439. https://doi.org/10.1002/cphc.201402473

Patel, A. R., & Dewettinck, K. (2015a). Comparative evaluation of structured oil systems: Shellac oleogel, HPMC oleogel, and HIPE gel. *European Journal of Lipid Science and Technology*, *117*(11), 1772–1781. https://doi.org/10.1002/ejlt.201400553

Patel, A. R., & Dewettinck, K. (2015b). Current update on the influence of minor lipid components, shear and presence of interfaces on fat crystallization. *Current Opinion in Food Science*, *3*, 65–70. https://doi.org/10.1016/j.cofs.2015.05.010

Patel, A. R., & Dewettinck, K. (2016). Edible oil structuring: An overview and recent updates. *Food & Function*, *7*(1), 20–29. https://doi.org/10.1039/c5fo01006c

Patel, A. R., Rajarethinem, P. S., Cludts, N., Lewille, B., De Vos, W. H., Lesaffer, A., & Dewettinck, K. (2015). Biopolymer-based structuring of liquid oil into soft solids and oleogels using water-continuous emulsions as templates. *Langmuir*, *31*(7), 2065–2073. https://doi.org/10.1021/la502829u

Patel, A. R., Schatteman, D., Lesaffer, A., & Dewettinck, K. (2013). A foam-templated approach for fabricating organogels using a water-soluble polymer. *Rsc Advances*, *3*(45), 22900–22903. https://doi.org/10.1039/c3ra44763d

Patterson, E., Wall, R., Fitzgerald, G., Ross, R., & Stanton, C. (2012). Health implications of high dietary omega-6 polyunsaturated fatty acids. *Journal of Nutrition and Metabolism*, *2012*, https://doi.org/10.1155/2012/539426

Petersen, K. D., Kleeberg, K. K., Jahreis, G., Busch-Stockfisch, M., & Fritsche, J. (2012). Comparison of analytical and sensory lipid oxidation parameters in conventional and high-oleic rapeseed oil. *European Journal of Lipid Science and Technology*, *114*(10), 1193–1203. https://doi.org/10.1002/ejlt.201200112

Phoon, P. Y., Paul, L. N., Burgner, J. W., San Martin-Gonzalez, M. F., & Narsimhan, G. (2014). Effect of cross-linking of interfacial sodium caseinate by natural processing on the oxidative stability of oil-in-water (O/W) emulsions. *Journal of Agricultural and Food Chemistry*, *62*(13), 2822–2829. https://doi.org/10.1021/jf403285z

Phuah, E. T., Tang, T. K., Lee, Y. Y., Choong, T. S. Y., Tan, C. P., & Lai, O. M. (2015). Review on the current state of diacylglycerol production using enzymatic approach. *Food and Bioprocess Technology*, *8*(6), 1169–1186. https://doi.org/10.1007/s11947-015-1505-0

Plazzotta, S., Calligaris, S., & Manzocco, L. (2019). Structure of oleogels from κ-carrageenan templates as affected by supercritical-CO_2-drying, freeze-drying and lettuce-filler addition. *Food Hydrocolloids*, *96*, 1–10. https://doi.org/10.1016/j.foodhyd.2019.05.008

Povey, M. J. (2017). Applications of ultrasonics in food science-novel control of fat crystallization and structuring. *Current Opinion in Colloid & Interface Science*, *28*, 1–6. https://doi.org/10.1016/j.cocis.2016.12.001

Qi, J. F., Wang, X. Y., Shin, J. A., Lee, Y. H., Jang, Y. S., Lee, J. H., … Lee, K. T. (2015). Relative oxidative stability of diacylglycerol

and triacylglycerol oils. *Journal of Food Science*, *80*(3), C510–C514. https://doi.org/10.1111/1750-3841.12792

Qiu, C. Y., Huang, Y., Li, A. J., Ma, D., & Wang, Y. (2018). Fabrication and characterization of oleogel stabilized by gelatin-polyphenol-polysaccharides nanocomplexes. *Journal of Agricultural and Food Chemistry*, *66*(50), 13243–13252. https://doi.org/10.1021/acs.jafc.8b02039

Rao, M. A. (2010). *Rheology of fluid and semisolid foods: Principles and applications*. Boston, MA: Springer. https://doi.org/10.1007/978-0-387-70930-7

Rao, R., Sankar, K. U., Sambaiah, K., & Lokesh, B. R. (2001). Differential scanning calorimetric studies on structured lipids from coconut oil triglycerides containing stearic acid. *European Food Research and Technology*, *212*(3), 334–343. https://doi.org/10.1007/s002170000254

Ribeiro, A. P. B., Basso, R. C., Grimaldi, R., Gioielli, L. A., & Gonçalves, L. a. G. (2009a). Instrumental methods for the evaluation of interesterified fats. *Food Analytical Methods*, *2*(4), 282–302. https://doi.org/10.1007/s12161-009-9073-4

Ribeiro, A. P. B., Grimaldi, R., Gioielli, L. A., & Gonçalves, L. A. (2009b). Zero *trans* fats from soybean oil and fully hydrogenated soybean oil: Physico-chemical properties and food applications. *Food Research International*, *42*(3), 401–410. https://doi.org/10.1016/j.foodres.2009.01.012

Rigolle, A., Gheysen, L., Depypere, F., Landuyt, A., Van Den Abeele, K., & Foubert, I. (2015). Lecithin influences cocoa butter crystallization depending on concentration and matrix. *European Journal of Lipid Science and Technology*, *117*(11), 1722–1732. https://doi.org/10.1002/ejlt.201400555

Rigolle, A., Van Den Abeele, K., & Foubert, I. (2018). Conventional and new techniques to monitor lipid crystallization. In K. Sato (Ed.), *Crystallization of lipids: Fundamentals and applications in food, cosmetics, and pharmaceuticals* (pp. 465–492). Hoboken, NJ: John Wiley & Sons. https://doi.org/10.1002/9781118593882.ch17

Rodriguez, B. a. M. (2019). Nonlinear rheology of fats using large amplitude oscillatory shear tests. In A. G. Marangoni (Ed.), *Structure-function analysis of edible fats* (pp. 169–195). Urbana, IL: AOCS Press. https://doi.org/10.1016/B978-0-12-814041-3.00006-X

Rohm, H., Schäper, C., & Zahn, S. (2018). Interesterified fats in chocolate and bakery products: A concise review. *LWT-Food Science and Technology*, *87*, 379–384. https://doi.org/10.1016/j.lwt.2017.08.076

Roman, O., Heyd, B., Broyart, B., Castillo, R., & Maillard, M.-N. (2013). Oxidative reactivity of unsaturated fatty acids from sunflower, high oleic sunflower and rapeseed oils subjected to heat treatment, under controlled conditions. *LWT-Food Science and Technology*, *52*(1), 49–59. https://doi.org/10.1016/j.lwt.2012.12.011

Saberi, A. H., Lai, O. M., & Toro-Vázquez, J. F. (2011). Crystallization kinetics of palm oil in blends with palm-based diacylglycerol. *Food Research International*, *44*(1), 425–435. https://doi.org/10.1016/j.foodres.2010.09.029

Saberi, A. H., Tan, C. P., & Lai, O. M. (2011). Phase behavior of palm oil in blends with palm-based diacylglycerol. *Journal of the American Oil Chemists' Society*, *88*(12), 1857–1865. https://doi.org/10.1007/s11746-011-1860-6

Saghafi, Z., Naeli, M. H., Tabibiazar, M., & Zargaraan, A. (2018). Zero-*trans* cake shortening: formulation and characterization of physicochemical, rheological, and textural properties. *Journal of the American Oil Chemists Society*, *95*(2), 171–183. https://doi.org/10.1002/aocs.12033

Şahin-Yeşilçubuk, N., & Akoh, C. C. (2017). Biotechnological and novel approaches for designing structured lipids intended for infant nutrition. *Journal of the American Oil Chemists' Society*, *94*(8), 1005–1034. https://doi.org/10.1007/s11746-017-3013-z

Saitou, K., Homma, R., Kudo, N., Katsuragi, Y., & Sato, K. (2014). Retardation of crystallization of diacylglycerol oils using polyglycerol fatty acid esters. *Journal of the American Oil Chemists' Society*, *91*(5), 711–719. https://doi.org/10.1007/s11746-014-2416-3

Saitou, K., Mitsui, Y., Shimizu, M., Kudo, N., Katsuragi, Y., & Sato, K. (2012). Crystallization behavior of diacylglycerol-rich oils produced from rapeseed oil. *Journal of the American Oil Chemists' Society*, *89*(7), 1231–1239. https://doi.org/10.1007/s11746-012-2014-1

Saitou, K., Taguchi, K., Homma, R., Shimizu, M., Yasunaga, K., Katsuragi, Y., … Sato, K. (2017). Retardation mechanism of crystallization of diacylglycerols resulting from the addition of polyglycerol fatty acid esters. *Crystal Growth & Design*, *17*(9), 4749–4756. https://doi.org/10.1021/acs.cgd.7b00676

Sakuno, M. M., Matsumoto, S., Kawai, S., Taihei, K., & Matsumura, Y. (2008). Adsorption and structural change of β-lactoglobulin at the diacylglycerol– water interface. *Langmuir*, *24*(20), 11483–11488. https://doi.org/10.1021/la8018277

Salvia-Trujillo, L., Decker, E. A., & Mcclements, D. J. (2016). Influence of an anionic polysaccharide on the physical and oxidative stability of omega-3 nanoemulsions: Antioxidant effects of alginate. *Food Hydrocolloids*, *52*, 690–698. https://doi.org/10.1016/j.foodhyd.2015.07.035

Sarkar, A., Horne, D. S., & Singh, H. (2010). Interactions of milk protein-stabilized oil-in-water emulsions with bile salts in a simulated upper intestinal model. *Food Hydrocolloids*, *24*(2-3), 142–151. https://doi.org/10.1016/j.foodhyd.2009.08.012

Sarkar, A., Li, H., Cray, D., & Boxall, S. (2018). Composite whey protein–cellulose nanocrystals at oil-water interface: Towards delaying lipid digestion. *Food Hydrocolloids*, *77*, 436–444. https://doi.org/10.1016/j.foodhyd.2017.10.020

Sarkar, A., Ye, A., & Singh, H. (2016). On the role of bile salts in the digestion of emulsified lipids. *Food Hydrocolloids*, *60*, 77–84. https://doi.org/10.1016/j.foodhyd.2016.03.018

Sarkar, A., Zhang, S., Holmes, M., & Ettelaie, R. (2019). Colloidal aspects of digestion of Pickering emulsions: Experiments and theoretical models of lipid digestion kinetics. *Advances in Colloid and Interface Science*, *263*, 195–211. https://doi.org/10.1016/j.cis.2018.10.002

Scholten, E. (2019). Edible oleogels: How suitable are proteins as a structurant? *Current Opinion in Food Science*, *27*, 36–42. https://doi.org/10.1016/j.cofs.2019.05.001

Sengupta, A., Roy, S., Mukherjee, S., & Ghosh, M. (2015). Production of medium chain fatty acid rich mustard oil using packed bed bioreactor. *Journal of Oleo Science*, *64*(2), 153–159. https://doi.org/10.5650/jos.ess14184

Shahidi, F. (2005). *Bailey's industrial oil & fats products*. Hoboken, NJ: John Wiley & Sons.

Shahidi, F., & Zhong, Y. (2010). Lipid oxidation and improving the oxidative stability. *Chemical Society Reviews*, *39*(11), 4067–4079. https://doi.org/10.1039/B922183M

Shen, Z., & Wijesundera, C. (2009). Effects of docosahexaenoic acid positional distribution on the oxidative stability of model triacylglycerol in water emulsion. *Journal of Food Lipids*, *16*(1), 62–71. https://doi.org/10.1111/j.1745-4522.2009.01132.x

Shimada, A., & Ohashi, K. (2003). Interfacial and emulsifying properties of diacylglycerol. *Food Science and Technology Research*, *9*(2), 142–147. https://doi.org/10.3136/fstr.9.142

Shimizu, M., Moriwaki, J., Nishide, T., & Nakajima, Y. (2004). Thermal deterioration of diacylglycerol and triacylglycerol oils during deep-frying. *Journal of the American Oil Chemists' Society*, *81*(6), 571–576. https://doi.org/10.1007/s11746-006-0943-2

Shin, J. A., Lee, M. Y., & Lee, K. T. (2016). Oxidation stability of O/W emulsion prepared with linolenic acid enriched diacylglycerol. *Journal of Food Science*, *81*(10), C2373–C2380. https://doi.org/10.1111/1750-3841.13421

Shrestha, L. K., Aramaki, K., Kato, H., Takase, Y., & Kunieda, H. (2006a). Foaming properties of monoglycerol fatty acid esters in nonpolar oil systems. *Langmuir*, *22*(20), 8337–8345. https://doi.org/10.1021/la061204h

Shrestha, L. K., Kaneko, M., Sato, T., Acharya, D. P., Iwanaga, T., & Kunieda, H. (2006b). Phase behavior of diglycerol fatty acid esters-nonpolar oil systems. *Langmuir*, *22*(4), 1449–1454. https://doi.org/10.1021/la052622+

Shrestha, L. K., Sato, T., Acharya, D. P., Iwanaga, T., Aramaki, K., & Kunieda, H. (2006c). Phase behavior of monoglycerol fatty acid esters in nonpolar oils: Reverse rodlike micelles at elevated temperatures. *The Journal of Physical Chemistry B*, *110*(25), 12266–12273. https://doi.org/10.1021/jp060587p

Shrestha, L. K., Shrestha, R. G., Sharma, S. C., & Aramaki, K. (2008). Stabilization of nonaqueous foam with lamellar liquid crystal particles in diglycerol monolaurate/olive oil system. *Journal of Colloid and Interface Science*, *328*(1), 172–179. https://doi.org/10.1016/j.jcis.2008.08.051

Shrestha, R. G., Shrestha, L. K., Solans, C., Gonzalez, C., & Aramaki, K. (2010). Nonaqueous foam with outstanding stability in diglycerol monomyristate/olive oil system. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *353*(2–3), 157–165. https://doi.org/10.1016/j.colsurfa.2009.11.007

Shuai, W., Das, R. K., Naghdi, M., Brar, S. K., & Verma, M. (2017). A review on the important aspects of lipase immobilization on nanomaterials. *Biotechnology and Applied Biochemistry*, *64*(4), 496–508. https://doi.org/10.1002/bab.1515

Sijtsma, L., & De Swaaf, M. (2004). Biotechnological production and applications of the ω-3 polyunsaturated fatty acid docosahexaenoic acid. *Applied Microbiology and Biotechnology*, *64*(2), 146–153. https://doi.org/10.1007/s00253-003-1525-y

Singh, A., Auzanneau, F. I., & Rogers, M. A. (2017). Advances in edible oleogel technologies—A decade in review. *Food Research International*, *97*, 307–317. https://doi.org/10.1016/j.foodres.2017.04.022

Singh, H., & Sarkar, A. (2011). Behaviour of protein-stabilised emulsions under various physiological conditions. *Advances in Colloid and Interface Science*, *165*(1), 47–57. https://doi.org/10.1016/j.cis.2011.02.001

Smith, K. W., Bhaggan, K., Talbot, G., & Van Malssen, K. F. (2011). Crystallization of fats: Influence of minor components and additives. *Journal of the American Oil Chemists' Society*, *88*(8), 1085–1101. https://doi.org/10.1007/s11746-011-1819-7

Smith, S. A., King, R. E., & Min, D. B. (2007). Oxidative and thermal stabilities of genetically modified high oleic sunflower oil. *Food Chemistry*, *102*(4), 1208–1213. https://doi.org/10.1016/j.foodchem.2006.06.058

Solaesa, G., Sanz, M. T., Falkeborg, M., Beltrán, S., & Guo, Z. (2016). Production and concentration of monoacylglycerols rich in omega-3 polyunsaturated fatty acids by enzymatic glycerolysis and molecular distillation. *Food Chemistry*, *190*, 960–967. https://doi.org/10.1016/j.foodchem.2015.06.061

Soumanou, M. M., Perignon, M., & Villeneuve, P. (2013). Lipase-catalyzed interesterification reactions for human milk fat substitutes production: A review. *European Journal of Lipid Science and Technology*, *115*(3), 270–285. https://doi.org/10.1002/ejlt.201200084

Souza, C. D., & Block, J. M. (2018). Impact of the addition of cocoa butter equivalent on the volatile compounds profile of dark chocolate. *Journal of Food Science and Technology-Mysore*, *55*(2), 767–775. https://doi.org/10.1007/s13197-017-2989-6

Speranza, P., & Macedo, G. A. (2012). Lipase-mediated production of specific lipids with improved biological and physicochemical properties. *Process Biochemistry*, *47*(12), 1699–1706. https://doi.org/10.1016/j.procbio.2012.07.006

Sproston, M. J., & Akoh, C. C. (2016a). Antioxidative effects of a glucose-cysteine maillard reaction product on the oxidative stability of a structured lipid in a complex food emulsion. *Journal of Food Science*, *81*(12), C2923–C2931. https://doi.org/10.1111/1750-3841.13541

Sproston, M. J., & Akoh, C. C. (2016b). Enzymatic modification of anhydrous milkfat with n-3 and n-6 fatty acids for potential use in infant formula: Comparison of methods. *Journal of the American Oil Chemists' Society*, *93*(2), 251–265. https://doi.org/10.1007/s11746-015-2763-8

Stortz, T. A., & Marangoni, A. G. (2011). Heat resistant chocolate. *Trends in food science & technology*, *22*(5), 201–214. https://doi.org/10.1016/j.tifs.2011.02.001

Tang, D., & Marangoni, A. G. (2007). Modeling the rheological properties and structure of colloidal fat crystal networks. *Trends in Food Science and Technology*, *18*(9), 474–483. https://doi.org/10.1016/j.tifs.2007.04.015

Tanti, R., Barbut, S., & Marangoni, A. G. (2016a). Hydroxypropyl methylcellulose and methylcellulose structured oil as a replacement for shortening in sandwich cookie creams. *Food Hydrocolloids*, *61*, 329–337. https://doi.org/10.1016/j.foodhyd.2016.05.032

Tanti, R., Barbut, S., & Marangoni, A. G. (2016b). Oil stabilization of natural peanut butter using food grade polymers. *Food Hydrocolloids*, *61*, 399–408. https://doi.org/10.1016/j.foodhyd.2016.05.034

Tavernier, I., Moens, K., Heyman, B., Danthine, S., & Dewettinck, K. (2019a). Relating crystallization behavior of monoacylglycerols-diacylglycerol mixtures to the strength of their crystalline network in oil. *Food Research International*, *120*, 504–513. https://doi.org/10.1016/j.foodres.2018.10.092

Tavernier, I., Norton, I. T., Rimaux, T., Lazidis, A., & Dewettinck, K. (2019b). Effect of high cooling and shear rate on the microstructural development of hybrid systems containing diacylglycerols and triacylglycerols of palm origin. *Journal of Food Engineering*, *246*, 141–152. https://doi.org/10.1016/j.jfoodeng.2018.11.009

Tavernier, I., Patel, A. R., Van Der Meeren, P., & Dewettinck, K. (2017). Emulsion-templated liquid oil structuring with soy protein and soy protein: Kappa-carrageenan complexes. *Food Hydrocolloids*, *65*, 107–120. https://doi.org/10.1016/j.foodhyd.2016.11.008

Tecelão, C., Perrier, V., Dubreucq, E., & Ferreira-Dias, S. (2019). Production of human milk fat substitutes by interesterification of tripalmitin with ethyl oleate catalyzed by *Candida parapsilosis* lipase/acyltransferase. *Journal of the American Oil Chemists' Society*, *96*(7), 777–787. https://doi.org/10.1002/aocs.12250

Teichert, S. A., & Akoh, C. C. (2011). Stearidonic acid soybean oil enriched with palmitic acid at the sn-2 position by enzymatic interesterification for use as human milk fat analogues. *Journal of Agricultural and Food Chemistry*, *59*(10), 5692–5701. https://doi.org/10.1021/jf200336t

Tian, F., Decker, E. A., Mcclements, D. J., & Goddard, J. M. (2014). Influence of non-migratory metal-chelating active packaging film on food quality: Impact on physical and chemical stability of emulsions. *Food Chemistry*, *151*, 257–265. https://doi.org/10.1016/j.foodchem.2013.11.074

Todokoro, T., Fukuda, K., Matsumura, K., Irie, M., & Hata, Y. (2016). Production of the natural iron chelator deferriferrichrysin from *Aspergillus oryzae* and evaluation as a novel food-grade antioxidant. *Journal of the Science of Food and Agriculture*, *96*(9), 2998–3006. https://doi.org/10.1002/jsfa.7469

Tran, T., & Rousseau, D. (2016). Influence of shear on fat crystallization. *Food Research International*, *81*, 157–162. https://doi.org/10.1016/j.foodres.2015.12.022

Truong, T., Prakash, S., & Bhandari, B. (2019). Effects of crystallisation of native phytosterols and monoacylglycerols on foaming properties of whipped oleogels. *Food Chemistry*, *285*, 86–93. https://doi.org/10.1016/j.foodchem.2019.01.134

Turan, D., SAhin YeşIlçUbuk, N. E., & Akoh, C. C. (2012). Production of human milk fat analogue containing docosahexaenoic and arachidonic acids. *Journal of Agricultural and Food Chemistry*, *60*(17), 4402–4407. https://doi.org/10.1021/jf3012272

Utama, Q. D., Sitanggang, A. B., Adawiyah, D. R., & Hariyadi, P. (2019). Lipase-catalyzed interesterification for the synthesis of medium-long-medium (MLM) structured lipids–A review. *Food Technology and Biotechnology*, *57*(3), 305–318. https://doi.org/10.17113/ftb.57.03.19.6025

Van Erp, H., Bryant, F. M., Martin-Moreno, J., Michaelson, L. V., Bhutada, G., & Eastmond, P. J. (2019). Engineering the stereoisomeric structure of seed oil to mimic human milk fat. *Proceedings of the National Academy of Sciences of the United States of America*, *116*(42), 20947–20952. https://doi.org/10.1073/pnas.1907915116

Vereecken, J., Meeussen, W., Foubert, I., Lesaffer, A., Wouters, J., & Dewettinck, K. (2009). Comparing the crystallization and polymorphic behaviour of saturated and unsaturated monoglycerides. *Food Research International*, *42*(10), 1415–1425. https://doi.org/10.1016/j.foodres.2009.07.006

Verstringe, S., Danthine, S., Blecker, C., Depypere, F., & Dewettinck, K. (2013). Influence of monopalmitin on the isothermal crystallization mechanism of palm oil. *Food Research International*, *51*(1), 344–353. https://doi.org/10.1016/j.foodres.2012.12.034

Verstringe, S., Danthine, S., Blecker, C., & Dewettinck, K. (2014). Influence of a commercial monoacylglycerol on the crystallization mechanism of palm oil as compared to its pure constituents. *Food Research International*, *62*, 694–700. https://doi.org/10.1016/j.foodres.2014.04.049

Villeneuve, P., Durand, E., & Decker, E. A. (2018). The need for a new step in the study of lipid oxidation in heterophasic systems. *Journal of Agricultural and Food Chemistry*, *66*, 8433–8434. https://doi.org/10.1021/acs.jafc.8b03603

Wagh, A., Birkin, P., & Martini, S. (2016). High-intensity ultrasound to improve physical and functional properties of lipids. *Annual Review of Food Science and Technology*, *7*, 23–41. https://doi.org/10.1146/annurev-food-041715-033112

Wagh, A., & Martini, S. (2017). Crystallization behavior of fats: Effects of processing conditions. In C. C. Akoh (Ed.), *Food lipids: Chemistry, nutrition, and biotechnology* (pp. 327–348). Boca Raton: CRC press.

Wang, J., Liu, X., Wang, X., Dong, T., Zhao, X., Zhu, D., … Wu, G. (2016). Selective synthesis of human milk fat-style structured triglycerides from microalgal oil in a microfluidic reactor packed with immobilized lipase. *Bioresource Technology*, *220*, 132–141. https://doi.org/10.1016/j.biortech.2016.08.023

Wang, X., Wang, X., Wang, W., Jin, Q., & Wang, X. (2018a). Synthesis of docosapentaenoic acid-enriched diacylglycerols by enzymatic glycerolysis of *Schizochytrium* sp. oil. *Bioresource Technology*, *262*, 278–283. https://doi.org/10.1016/j.biortech.2018.04.061

Wang, X., Zou, S., Miu, Z., Jin, Q., & Wang, X. (2019). Enzymatic preparation of structured triacylglycerols with arachidonic and palmitic acids at the sn-2 position for infant formula use. *Food Chemistry*, *283*, 331–337. https://doi.org/10.1016/j.foodchem.2019.01.057

Wang, X. S., Wang, X. H., Wang, W., Jin, Q. Z., & Wang, X. G. (2018b). Synthesis of docosapentaenoic acid-enriched diacylglycerols by enzymatic glycerolysis of Schizochytrium sp oil. *Bioresource Technology*, *262*, 278–283. https://doi.org/10.1016/j.biortech.2018.04.061

Wang, Y., Zhao, M., Tang, S., Song, K., Han, X., & Ou, S. (2010). Evaluation of the oxidative stability of diacylglycerol-enriched soybean oil and palm olein under rancimat-accelerated oxidation conditions. *Journal of the American Oil Chemists' Society*, *87*(5), 483–491. https://doi.org/10.1007/s11746-009-1521-1

Waraho, T., Mcclements, D. J., & Decker, E. A. (2011). Mechanisms of lipid oxidation in food dispersions. *Trends in Food Science and Technology*, *22*(1), 3–13. https://doi.org/10.1016/j.tifs.2010.11.003

Wei, W., Jin, Q. Z., & Wang, X. G. (2019). Human milk fat substitutes: Past achievements and current trends. *Progress in Lipid Research*, *74*, 69–86. https://doi.org/10.1016/j.plipres.2019.02.001

West, R., & Rousseau, D. (2017). Modelling sugar, processing, and storage effects on palm oil crystallization and rheology. *LWT-Food Science and Technology*, *83*, 201–212. https://doi.org/10.1016/j.lwt.2017.05.021

Wijaya, W., Sun, Q. Q., Vermeir, L., Dewettinck, K., Patel, A. R., & Van Der Meeren, P. (2019). pH and protein to polysaccharide ratio control the structural properties and viscoelastic network of HIPE-templated biopolymeric oleogels. *Food Structure-Netherlands*, *21*, 100112, https://doi.org/Unsp10011210.1016/J.Foostr.2019.100112

Wijesundera, C. (2008). The influence of triacylglycerol structure on the oxidative stability of polyunsaturated oils. *Lipid Technology*, *20*(9), 199–202. https://doi.org/10.1002/lite.200800051

Wijesundera, C., Ceccato, C., Watkins, P., Fagan, P., Fraser, B., Thienthong, N., & Perlmutter, P. (2008). Docosahexaenoic acid is more stable to oxidation when located at the sn-2 position of triacylglycerol compared to sn-1 (3). *Journal of the American Oil Chemists' Society*, *85*(6), 543–548. https://doi.org/10.1007/s11746-008-1224-z

Wilde, P. J., & Chu, B. S. (2011). Interfacial & colloidal aspects of lipid digestion. *Advances in Colloid and Interface Science*, *165*(1), 14–22. https://doi.org/10.1016/j.cis.2011.02.004

Wilkes, R. S. (2008). Low linolenic soybeans and beyond. *Lipid Technology*, *20*(12), 277–279. https://doi.org/10.1002/lite.200800072

Willett, S. A., & Akoh, C. C. (2019a). Physicochemical characterization of organogels prepared from menhaden oil or structured lipid with phytosterol blend or sucrose stearate/ascorbyl palmitate blend. *Food & Function*, *10*(1), 180–190. https://doi.org/10.1039/c8fo01725e

Willett, S. A., & Akoh, C. C. (2019b). Physicochemical characterization of yellow cake prepared with structured lipid oleogels. *Journal of Food Science*, *84*(6), 1390–1399. https://doi.org/10.1111/1750-3841.14624

Winuprasith, T., Khomein, P., Mitbumrung, W., Suphantharika, M., Nitithamyong, A., & Mcclements, D. J. (2018). Encapsulation of vitamin D_3 in pickering emulsions stabilized by nanofibrillated mangosteen cellulose: Impact on *in vitro* digestion and bioaccessibility. *Food Hydrocolloids*, *83*, 153–164. https://doi.org/10.1016/j.foodhyd.2018.04.047

Wright, A. J., Hartel, R. W., Narine, S. S., & Marangoni, A. G. (2000). The effect of minor components on milk fat crystallization. *Journal of the American Oil Chemists Society*, *77*(5), 463–475. https://doi.org/https://doi.org/10.1007/s11746-000-0075-8

Wu, G., Zaniolo, O., Schuster, H., Schlotzer, E., & Pradelli, L. (2017). Structured triglycerides versus physical mixtures of medium-and long-chain triglycerides for parenteral nutrition in surgical or critically ill adult patients: Systematic review and meta-analysis. *Clinical Nutrition*, *36*(1), 150–161. https://doi.org/10.1016/j.clnu.2016.01.004

Xia, Q. Y., Akanbi, T. O., Li, R., Wang, B., Yang, W. R., & Barrow, C. J. (2019). Lipase-catalysed synthesis of palm oil-omega-3 structured lipids. *Food & Function*, *10*(6), 3142–3149. https://doi.org/10.1039/c9fo00668k

Xiao, Y., Zheng, M., Liu, Z., Shi, J., Huang, F., & Luo, X. (2018). Constructing a continuous flow bioreactor based on a hierarchically porous cellulose monolith for ultrafast and non-stop enzymatic esterification/transesterification. *ACS Sustainable Chemistry & Engineering*, *7*(2), 2056–2063. https://doi.org/10.1021/acssuschemeng.8b04471

Xie, W., & Zang, X. (2017). Covalent immobilization of lipase onto aminopropyl-functionalized hydroxyapatite-encapsulated-γ-Fe_2O_3 nanoparticles: A magnetic biocatalyst for interesterification of soybean oil. *Food Chemistry*, *227*, 397–403. https://doi.org/10.1016/j.foodchem.2017.01.082

Xie, W., & Zang, X. (2018). Lipase immobilized on ionic liquid-functionalized magnetic silica composites as a magnetic biocatalyst for production of *trans*-free plastic fats. *Food Chemistry*, *257*, 15–22. https://doi.org/10.1016/j.foodchem.2018.03.010

Xu, Y., & Cao, D. (2017a). Phase behavior of binary mixtures of three different 1,3-diacylglycerols. *European Journal of Lipid Science and Technology*, *119*(9), 1600497. https://doi.org/10.1002/ejlt.201600497

Xu, Y., & Cao, D. (2017b). Thermal and kinetic behaviors and microscopic characteristics of diacylglycerol-enriched palm-based oils blends. *European Journal of Lipid Science and Technology*, *119*(9), 1600491. https://doi.org/10.1002/ejlt.201600491

Xu, Y., Guo, S., Wang, W., Wang, Y., & Yang, B. (2013). Enzymatic hydrolysis of palm stearin to produce diacylglycerol with a highly thermostable lipase. *European Journal of Lipid Science and Technology*, *115*(5), 564–570. https://doi.org/10.1002/ejlt.201200373

Xu, Y., Wei, C., Zhao, X., Chen, L., & Cao, D. (2016). A comparative study on microstructure, texture, rheology, and crystallization kinetics of palm-based diacylglycerol oils and corresponding palm-based oils. *European Journal of Lipid Science and Technology*, *118*(8), 1179–1192. https://doi.org/10.1002/ejlt.201500369

Xu, Y., Zhu, X., Ma, X., Xiong, H., Zeng, Z., Peng, H., & Hu, J. (2018). Enzymatic production of *trans*-free shortening from coix seed oil, fully hydrogenated palm oil and *Cinnamomum camphora* seed oil. *Food Bioscience*, *22*, 1–8. https://doi.org/10.1016/j.fbio.2017.12.010

Yamoneka, J., Malumba, P., Lognay, G., Béra, F., Blecker, C., & Danthine, S. (2018b). Enzymatic inter-esterification of binary blends containing *Irvingia gabonensis* seed fat to produce cocoa butter substitute. *European Journal of Lipid Science and Technology*, *120*(4), 1700423. https://doi.org/10.1002/ejlt.201700423

Yanai, H., Tomono, Y., Ito, K., Furutani, N., Yoshida, H., & Tada, N. (2007). Diacylglycerol oil for the metabolic syndrome. *Nutrition Journal*, *6*(1), 43. https://doi.org/10.1186/1475-2891-6-43

Ye, X. Y., Li, P. X., Lo, Y. M., Fu, H., & Cao, Y. P. (2019). Development of novel shortenings structured by ethylcellulose oleogels. *Journal of Food Science*, *84*(6), 1456–1464. https://doi.org/10.1111/1750-3841.14615

Yeoh, C. M., Phuah, E. T., Tang, T. K., Siew, W. L., Abdullah, L. C., & Choong, T. S. Y. (2014). Molecular distillation and characterization of diacylglycerol-enriched palm olein. *European Journal of Lipid Science and Technology*, *116*(12), 1654–1663. https://doi.org/10.1002/ejlt.201300502

Yesiltas, B., Sørensen, A. D. M., García-Moreno, P. J., Anankanbil, S., Guo, Z., & Jacobsen, C. (2019). Modified phosphatidylcholine with different alkyl chain length and covalently attached caffeic acid affects the physical and oxidative stability of omega-3 delivery 70% oil-in-water emulsions. *Food Chemistry*, *289*, 490–499. https://doi.org/10.1016/j.foodchem.2019.03.087

Zam, W. (2015). Structured lipids: Methods of production, commercial products and nutraceutical characteristics. *Progress in Nutrition*, *17*(3), 198–213.

Zare, D., Allison, J. R., & Mcgrath, K. M. (2016). Molecular dynamics simulation of β-lactoglobulin at different oil/water interfaces. *Biomacromolecules*, *17*(5), 1572–1581. https://doi.org/10.1021/acs.biomac.5b01709

Zhai, J., Wooster, T. J., Hoffmann, S. V., Lee, T. H., Augustin, M. A., & Aguilar, M. I. (2011). Structural rearrangement of β-lactoglobulin at different oil–water interfaces and its effect on emulsion stability. *Langmuir*, *27*(15), 9227–9236. https://doi.org/10.1021/la201483y

Zhang, K., Wang, W. H., Wang, X., Cheng, S., Zhou, J. Y., Wu, Z. A., & Li, Y. (2019). Fabrication and physicochemical and antibacterial properties of ethyl cellulose-structured cinnamon oil oleogel: Relation between ethyl cellulose viscosity and oleogel performance. *Journal of the Science of Food and Agriculture*, *99*(8), 4063–4071. https://doi.org/10.1002/jsfa.9636

Zhang, N., Yang, X., Fu, J., Chen, Q., Song, Z., & Wang, Y. (2016). Production of diacylglycerol-enriched oil by glycerolysis of soybean oil using a bubble column reactor in a solvent-free system. *Journal of Oleo Science*, *65*(3), 207–216. https://doi.org/10.5650/jos.ess15241

Zhang, Y., Wang, X., Xie, D., Zou, S., Jin, Q., & Wang, X. (2018a). Synthesis and concentration of 2-monoacylglycerols rich in polyunsaturated fatty acids. *Food Chemistry*, *250*, 60–66. https://doi.org/10.1016/j.foodchem.2018.01.027

Zhang, Y., Wang, X., Zou, S., Xie, D., Jin, Q., & Wang, X. (2018b). Synthesis of 2-docosahexaenoylglycerol by enzymatic ethanolysis. *Bioresource Technology*, *251*, 334–340. https://doi.org/10.1016/j.biortech.2017.12.025

Zhao, J., Wang, T., Lin, J., Yang, L., & Wu, M. (2019). Preparation of high-purity 1,3-diacylglycerol using performance-enhanced lipase immobilized on nanosized magnetite particles. *Biotechnology and Bioprocess Engineering*, *24*(2), 326–336. https://doi.org/10.1007/s12257-018-0458-3

Zhao, X., Sun, Q., Qin, Z., Liu, Q., & Kong, B. (2018). Ultrasonic pretreatment promotes diacylglycerol production from lard by lipase-catalysed glycerolysis and its physicochemical properties. *Ultrasonics Sonochemistry*, *48*, 11–18. https://doi.org/10.1016/j.ultsonch.2018.05.005

Zheng, M., Wang, S., Xiang, X., Shi, J., Huang, J., Deng, Q., … Xiao, J. (2017). Facile preparation of magnetic carbon nanotubes-immobilized lipase for highly efficient synthesis of 1,3-dioleoyl-2-palmitoylglycerol-rich human milk fat substitutes. *Food Chemistry*, *228*, 476–483. https://doi.org/10.1016/j.foodchem.2017.01.129

Zhong, N., Chen, W., Liu, L., & Chen, H. (2019). Immobilization of *Rhizomucor miehei* lipase onto the organic functionalized SBA-15: Their enzymatic properties and glycerolysis efficiencies for diacylglycerols production. *Food Chemistry*, *271*, 739–746. https://doi.org/10.1016/j.foodchem.2018.07.185

Zhu, T., Weng, H., Zhang, X., Wu, H., & Li, B. (2018a). Mechanistic insight into the relationship between triacylglycerol and crystallization of lipase-catalyzed interesterified blend of palm stearin and vegetable oil. *Food Chemistry*, *260*, 306–316. https://doi.org/10.1016/j.foodchem.2018.03.147

Zhu, T., Zhang, X., Wu, H., & Li, B. (2019). Comparative study on crystallization behaviors of physical blend-and interesterified blend-based special fats. *Journal of Food Engineering*, *241*, 33–40. https://doi.org/10.1016/j.jfoodeng.2018.08.005

Zhu, T. W., Liu, Q., Weng, H. T., Li, B., Zhang, X., Zong, M. H., & Wu, H. (2018b). Effect of temperature on the crystallization behavior and physical properties of fast-frozen special fat during storage. *Journal of Food Engineering*, *223*, 53–61. https://doi.org/10.1016/j.jfoodeng.2017.12.002

Zhu, T. W., Zhao, Y. L., Zong, M. H., Li, B., Zhang, X., & Wu, H. (2017). Improvement of physical properties of palm stearin and soybean oil blends by enzymatic interesterification and their application in fast frozen food. *RSC Advances*, *7*(55), 34435–34441. https://doi.org/10.1039/c7ra02829f

Zou, L., & Akoh, C. C. (2015a). Antioxidant activities of annatto and palm tocotrienol-rich fractions in fish oil and structured lipid-based infant formula emulsion. *Food Chemistry*, *168*, 504–511. https://doi.org/10.1016/j.foodchem.2014.07.098

Zou, L., & Akoh, C. C. (2015b). Oxidative stability of structured lipid-based infant formula emulsion: Effect of antioxidants. *Food Chemistry*, *178*, 1–9. https://doi.org/10.1016/j.foodchem.2015.01.073

Zou, L., Pande, G., & Akoh, C. C. (2016a). Infant formula fat analogs and human milk fat: New focus on infant developmental needs. *Annual Review of Food Science and Technology*, *7*(1), 139–165. https://doi.org/10.1146/annurev-food-041715-033120

Zou, X., Jin, Q., Guo, Z., Xu, X., & Wang, X. (2016b). Preparation of human milk fat substitutes from basa catfish oil: Combination of enzymatic acidolysis and modeled blending. *European Journal of Lipid Science and Technology*, *118*(11), 1702–1711. https://doi.org/10.1002/ejlt.201500591

Zulkurnain, M., Maleky, F., & Balasubramaniam, V. (2016). High pressure processing effects on lipids thermophysical properties and crystallization kinetics. *Food Engineering Reviews*, *8*(4), 393–413. https://doi.org/10.1007/s12393-016-9144-4

How to cite this article: Guo Y, Cai Z, Xie Y, et al. Synthesis, physicochemical properties, and health aspects of structured lipids: a review. *Compr Rev Food Sci Food Saf*. 2020;19:759–800. https://doi.org/10.1111/1541-4337.12537