

Monoacylglycerols and diacylglycerols for fat-based food products: a review

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Abstract

The flexibility of monoacylglycerol (MAG) and diacylglycerol (DAG) attracted the attention of researchers to learn more about MAG and DAG in food science and technology. The objective of this paper is to serve a review or summarize the production (especially enzymatic glycerolysis), characteristics, and application of MAG and DAG, especially for fat-based products. The characteristic of MAG and DAG related to melting and crystallization properties, polymorphism, microstructure, and rheology are presented and discussed. The applications of MAG and DAG include their use as fats/oils rich in MAG and DAG, margarine/shortening, surfactant/emulsifier, and organogels are also provided. Various studies have shown that MAG and DAG can improve the characteristics of various fat-based food products.

1. Introduction

Structured lipids (SLs) are lipids that have been modified in the composition of fatty acids or the distribution of fatty acid positions through chemical or enzymatic reactions. The scope of "lipids" includes acylglycerols, such as triacylglycerols (TAGs), diacylglycerols (DAGs), monoacylglycerols (MAGs) and phospholipids (Kim and Akoh, 2015). Research on SLs is growing rapidly, including monoacylglycerol and diacylglycerol (MAG and DAG) due to their nature as a versatile emulsifier. MAG and DAG can also improve the physical and functional characteristics of lipids, including crystallization and melting properties, solid fat content, texture and rheology, microstructure, and polymorphism (Saber *et al.*, 2012; Chen *et al.*, 2014; Ng *et al.*, 2014). MAG and DAG can also improve the oxidative properties of fats/oils (Gomes *et al.*, 2010; Nderitu *et al.*, 2018). MAG and DAG, especially in the form of 1,3-DAG, also have better nutritional value than TAG (Lo *et al.*, 2008; Feltes *et al.*, 2013).

MAG and DAG can be synthesized through partial hydrolysis of fat/oil (TAG), partial esterification of free fatty acids with glycerol, and glycerolysis of fats/oils with glycerol (Gunstone, 1999; Devi *et al.*, 2008; Feltes *et al.*, 2013). Partial hydrolysis has difficulty in controlling the hydrolysis reaction; yields are relatively low and require large amounts of oils/fats. Partial esterification between glycerol and free fatty acids is

another strategy to obtain MAG and DAG, but this reaction is reversible for re-hydrolysis. However, MAG and DAG are produced more effectively by glycerolysis between oil/fat (TAG) and glycerol. Glycerol can be obtained easily and abundantly from the byproducts of biodiesel processing in the form of glycerin so that the production of MAG and DAG through glycerolysis process is more attractive because of the cheapness of raw materials (Phuah *et al.*, 2015). Glycerolysis produces high yields because each mole of TAG can produce 3 moles of MAG. Glycerolysis can also be directed to the formation of DAG by reducing glycerol as a substrate (Bornscheuer, 1995; Subroto *et al.*, 2019).

MAG and DAG have been widely used in food, such as healthy oil-rich in DAG (Lo *et al.*, 2008; Ferretti *et al.*, 2018), shortening (Latip *et al.*, 2013), *soft tub margarine* (Saber *et al.*, 2012), organogels (Yilmaz and Ögütçü, 2014; Lopez-Martínez *et al.*, 2015), and *fatty emulsion* (Kowalska *et al.*, 2014). MAG and DAG are also widely used as an emulsifier in various emulsion-based products. Besides, they can improve polymorphic transitions, susceptibility to fat bloom, emulsion stability, and oxidation (Schantz and Rohm, 2005; Smith *et al.*, 2011). The utilization of MAG and DAG continues to develop in the application of fat/oil products with special properties by modifying the source of fats/oils and the types of fatty acids.

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2. Methods for MAG and DAG production

Of all the methods for producing MAG and DAG, glycerolysis is the method that theoretically gives the highest yield (Feltés *et al.*, 2013). Glycerolysis can also utilize glycerin which is a biodiesel industry waste (Ferrão *et al.*, 2017). On an industrial scale, MAG and DAG are generally produced through continuous glycerolysis of fat/oil using alkaline inorganic catalysts, high temperature (220-260°C), and an inert gas (Damstrup *et al.*, 2005).

MAG and DAG production can also be carried out with enzymatic catalysis, which gets attention because it does not require high temperatures (mild reaction), the lipids are not easily degraded, and the products are easily purified and are environmentally friendly. The lipase selectivity property also has the potential to obtain suitable products such as structured lipids (Gunstone, 1999). Lipase immobilization with chemical or physical modification can also increase its catalytic and can be easily separated and reused to reduce production costs and become a competitive biocatalyst to be applied to the industry (Villeneuve *et al.*, 2000; Osborn and Akoh, 2002).

MAG products or DAGs that are obtained subsequently need to be separated or purified, including through separation based on the formation of a solid phase resulting from glycerolysis. Melting points generally increase TAG, DAG, and MAG sequentially and decrease with increasing unsaturation of the fatty acids making up acylglycerol (Pawongrat *et al.*, 2008). Product separation can be carried out by fractionation at cold temperatures so that lipids with high melting points will crystallize and produce a solid phase. Separation of the solid fraction will separate the acylglycerols obtained. Separation or purification of MAG or DAG can also use molecular distillation, which is especially suitable for separating and purifying heat-sensitive and liquid-shaped products at low air pressure. This method can produce MAG with a purity of up to 90% (Corma *et al.*, 2007; Wang *et al.*, 2010).

3. Enzymatic glycerolysis

Enzymatic glycerolysis has the advantage that the reaction temperature is relatively low, the substrate is not degraded, purification of the product is easier and more environmentally friendly than chemical glycerolysis (Phuah *et al.*, 2015). Enzymatic glycerolysis only requires small amounts of water. Therefore, enzymes should be in an immobilized form. Besides, immobilized enzymes can be used repeatedly and have better stability (Osborn and Akoh, 2002). The enzymatic glycerolysis reaction is affected by several factors. These factors

include the type of stirrer in the reactor (Phuah *et al.*, 2015; Subroto *et al.*, 2020). Stirrer serves to accelerate the occurrence of enzyme contact with the substrate. Fat/oil and glycerol substrate do not dissolve each other. Thus a solvent is needed to dissolve it. Glycerol used as a substrate can increase the rate of the reaction but is too large a number to inhibit the reaction (Krüger *et al.*, 2010). The amount of enzyme/catalyst used can also accelerate the reaction to a certain concentration (Naik *et al.*, 2014; Subroto *et al.*, 2019).

Enzymatic glycerolysis can be directed to the formation of monoacylglycerol (MAG) and diacylglycerol (DAG), mainly by determining the amount of glycerol. Glycerolysis leads to the formation of MAG in excess glycerol conditions, whereas the reaction leads to the forming of DAG when the amount of glycerol is limited. Naik *et al.* (2014) investigated the glycerolysis of sunflower oil for MAG production obtaining an optimum amount of glycerol used at a oil to glycerol molar ratio of 1:5, whereas Wang *et al.* (2011) producing DAG through enzymatic glycerolysis of soybean oil obtained optimum conditions of oil to glycerol molar ratio of about 2:3.

In the process of glycerolysis using immobilized lipase therein consists of a hydrophobic component (oil/fat), hydrophilic component (glycerol), and an enzyme in the form of a solid phase. In such systems, it is imperative to pay attention to the low solubility between the substrate (glycerol and fat/oil) (Pawongrat *et al.*, 2007). Nevertheless, MAG and DAG obtained during the reaction can act as emulsifiers, increase the mixing between the two substrates and increase conversion rate. Under these conditions, glycerolysis can be carried out without the addition of solvents or other additives, but the results obtained are less than optimal due to the low solubility of the two-phase system (Torres *et al.*, 2002). An alternative way to disperse immiscible substrates is the use of surfactants, which are molecules that have amphipathic characteristics to form micelle systems. The use of surfactants in the glycerolysis system, followed by rapid stirring, can create an interfacial area, increasing contact between the substrate and enzymes, thereby increasing MAG and DAG products (Bornscheuer, 1995).

Enzymatic glycerolysis can also be conducted in various reaction systems, such as by forming micelles. The reaction is slower when glycerolysis is undertaken on a solvent-free system (Yang *et al.*, 2003; Valério *et al.*, 2009). These problems can be overcome by using organic solvents or on systems containing surface-active materials or surfactants (Yang *et al.*, 2005; Pawongrat *et al.*, 2007). The use of surfactants or solvents as reaction

media will reduce reaction time and increase conversion, so it is potential for industrial-scale application (Feltes *et al.*, 2013).

The alternative to overcome the low substrate solubility in glycerolysis is to use a suitable solvent and produce a homogeneous system that increases mass transfer, decreases reaction time, and increases product conversion. In such systems, t-butanol are extensively used because they produce high products in a short time. Naik *et al.* (2014) investigated glycerolysis of sunflower oil for the production of monoacylglycerol with the lipase catalyst *C. antarctica* using several types of organic solvents and found that t-butanol is the best solvent. Wang *et al.* (2011), on the other hand, examined the glycerolysis of soybean oil for the production of diacylglycerol using *C. antarctica* lipase in the t-butanol system obtained by the substrate concentration in the optimum t-butanol solvent at a concentration of 40%.

In general, the reaction media are critical in enzymatic glycerolysis because they can overcome the low miscibility of the substrates. The use of solvents makes the reaction system homogeneous, decreases the viscosity of the reaction mixture, increases the diffusion of the substrate, and increases the reaction rate (Tripathi *et al.*, 2006; Subroto *et al.*, 2019). Several studies have shown that differences in solvent polarity also affect lipase selectivity in catalyzing reactions. Some organic solvents provide a protective effect on lipase so that it is more stable to heat (Goldberg *et al.*, 1990).

The catalytic activity of lipases is efficient in organic solvents. When an enzyme is in an organic environment, its properties can change like its chemo- and stereoselectivity, increase its stability, and increase its rigidity. The higher the amount of the solvent, the higher the mass transfer and the lower the viscosity of the reactants (Naik *et al.*, 2014). The selection of organic solvents must pay attention to the solubility of the substrates in the solvent, and it has to be inert in the reaction. Other factors that may need to be considered are density, toxicity, viscosity, surface tension, and cost (Feltes *et al.*, 2013). Naik *et al.* (2014) found that for enzymatic glycerolysis between oil and glycerol, t-butanol and t-pentanol are better solvents than hexane, heptane, acetone, ethanol, methanol, acetonitrile, and chloroform.

Lipase from *C. antarctica B* (CALB) is an excellent biocatalyst for the glycerolysis reaction. This enzyme catalyzes a variety of different substrates and shows good performance (Torres *et al.*, 2002). CALB has excellent activity in the presence of solvents and surfactants (Feltes *et al.*, 2013). The use of CALB for the glycerolysis reaction is almost the same as the

interesterification reaction in general, which is 5-15%. Naik *et al.* (2014) reported that glycerolysis of sunflower oil for monoacylglycerol production obtained the optimum conditions at a CALB concentration of 15%. Subroto *et al.* (2019) found that glycerolysis of palm stearin and palm olein mixture using CALB was optimal at a concentration of 15%.

Lipase concentration can increase the rate of glycerolysis reaction, but the use of enzymes at concentrations that are too high can inhibit the reaction, especially if using an immobilized lipase in the form of solids. Naik *et al.* (2014) found that high enzyme concentrations could cause the active site of enzyme molecules not to be exposed to the substrate. Besides, protein aggregation tends to occur easily. Immobilized lipases in large quantities also cause less effective agitation, which affects the limitations of mass transfer (Krüger *et al.*, 2010; Subroto *et al.*, 2019). Another problem of enzymatic glycerolysis is the forming of a hydrophilic glycerol layer that envelops an immobilized lipase using a hydrophilic matrix, thus inhibiting contact with the hydrophobic oil phase (Kristensen *et al.*, 2005). It is necessary to choose an enzyme that is hydrophobic matrix so that it can catalyze glycerolysis optimally and obtain more product conversion (Phuah *et al.*, 2015).

Enzymatic glycerolysis is affected by various factors such as the type and amount of substrates, temperature, the use of solvents, and catalyst concentration. Substrate composition affects the rate of glycerolysis using lipases. Increasing the amount of substrate does not always increase the rate of the reaction. The use of an excessive amount of substrate will occur in substrate inhibition. In the glycerolysis reaction, the composition or substrate ratio between oil and glycerol affected the lipase activity, reaction rate, and product conversion. Naik *et al.* (2014), Wang *et al.* (2011), and Subroto *et al.* (2019) found that the addition of glycerol increased the rate of reaction and led to MAG formation rather than DAG, but excessive glycerol addition inhibited the conversion of TAG to MAG and DAG.

The glycerolysis reaction is also affected by the reaction temperature. Increasing the reaction temperature will increase the reaction rate, but too high a temperature can decrease the reaction rate and cause denaturation of the enzyme (Willis and Marangoni, 2002). The high temperature is not needed in a system that contains organic solvents because the hydrophobic substrate dissolves easily. Enzymes are proteins whose stability is related to the three-dimensional structure of proteins. The integrity of the three-dimensional structure on the active side is very important to maintain enzyme activity, and integrity is very much affected by the reaction

temperature (Whitaker, 1994). Immobilization maintains lipases in conformation, which decreases the ease of enzymes denatured by heat. The optimal temperature for most immobilized lipases is 30-62°C. Subroto *et al.* (2008) obtained that the optimum temperature of enzymatic acidolysis of fish oil with lauric acid is 40-50°C. Naik *et al.* (2014) and Wang *et al.* (2011), who carried out enzymatic glycerolysis using immobilized lipase from *C. antarctica* as a catalyst, obtained that the optimal temperature was reached at 50°C.

4. The characteristics of monoacylglycerol and diacylglycerol

MAG and DAG have different properties compared to TAG. Some characteristics such as melting and crystallization, polymorphism, microstructure, and rheology of a fat/oil are affected by the presence of MAG and DAG, as shown in Table 1.

4.1 Crystallization and melting characteristic

The minor lipid components, including MAG and DAG, affect the melting and crystallization characteristics of fats/oils (Smith *et al.*, 2011). These thermal characteristics are usually analyzed by differential scanning calorimeter (DSC) and are shown by a thermogram profile. MAG has faster crystal formation than DAG, and DAG has faster crystal formation than TAG (Basso *et al.*, 2010; Normah *et al.*, 2014). Fats/oils that have MAG and DAG at high concentrations or are added from the outside display more peaks of exothermic transitions, where the peaks are at a higher temperature than native fats/oils. Ng *et al.* (2014) and Saberi *et al.* (2012) reported that differences in the crystallization profiles were mainly due to the content of different acylglycerols structures (MAG, DAG, and TAG). MAG and DAG also have higher heats of crystallization (ΔH) than TAG. Saberi *et al.* (2011) compared palm-based oils and palm-based diacylglycerol oils. It was found that ΔH palm-based diacylglycerol was higher in all fractions. Crystallization

characteristics can further affect the texture of fat. Increasing the MAG and DAG content in fat accelerates the process of crystal formation and creates a harder texture (Basso *et al.*, 2010; Saberi, Tan and Lai, 2011).

Generally, MAG and DAG have a higher melting point than TAG. Saberi, Tan and Lai (2011) showed that in palm oil added to palm oil-DAG (PO-DAG), the peak endotherm of DAG increased in intensity and the peak shifted to a higher temperature. The results showed that palm oil added with palm-based diacylglycerols (P-DAG) had more melting peaks and at higher temperatures. Therefore, MAG and DAG can be utilized for the formation of fats/oils products that have faster crystallization and higher melting points than their natural fats/oils.

4.2 Solid fat content

In general, MAG and DAG have higher melting points and have implications for increasing SFC. MAG and DAG have SFC profiles with slower SFC declines than TAG and experience perfect melting (SFC = 0) at temperatures higher than TAG. Saberi *et al.* (2011) compared the SFC profile of palm-based diacylglycerols in various types of fractions with palm-based oils. It was found that the SFC profile in palm-based diacylglycerol has a higher SFC than palm-based oils especially at temperatures above 15°C. SFC in rich-in fats MAG or DAG which was gradually melted shows a relatively more uniform consistency at various temperatures, making it more suitable for plastic-fat products such as margarine and shortening (Nor Aini *et al.*, 2002). Romain *et al.* (2013) obtained monoacylglycerols from *Nephelium lappaceum L.* oil, showing that MAG fractions at various purification levels had significantly higher SFC at all temperatures. Naderi *et al.* (2016) reported that the addition of MAG (monopalmitin) above 0.5% increased the SFC of structured chicken fat at all temperatures. This shows that increasing the proportion of MAG results in the formation of a crystalline network that is more resistant to temperature; this can be

Table 1. Changes in MAG and DAG characteristics compared to TAG

Characteristics	Changes in characteristics	References
Crystallization	- Faster crystallization - Higher temperature	(Basso <i>et al.</i> , 2010; Saberi <i>et al.</i> , 2011; Normah <i>et al.</i> , 2014)
Melting	- Slower Melting - Higher temperature	(Basso <i>et al.</i> , 2010; Saberi <i>et al.</i> , 2011)
Solid fat content	- Higher solid fat content	(Saberi <i>et al.</i> , 2012; Ng <i>et al.</i> , 2014)
Polymorphism	- β -crystal \gg β' -crystal	(Saberi, Tan and Lai, 2011; Saberi <i>et al.</i> , 2011; Zhang <i>et al.</i> ,
Microstructure	- Smaller crystal - More homogeneous - Needle-like crystals	(Maruyama <i>et al.</i> , 2014; Xu <i>et al.</i> , 2016; Naderi <i>et al.</i> , 2016)
Rheology	- Higher viscosity and elasticity	(Rocha-Amador <i>et al.</i> , 2014; Naderi <i>et al.</i> , 2018)
Texture (Hardness)	- Harder	(Saberi, Tan and Lai, 2011; Subroto <i>et al.</i> , 2019)

attributed to the fat crystal structure which is denser at the addition of high MAG (Basso *et al.*, 2010; Maruyama *et al.*, 2014). Ng *et al.* (2014) studied palm olein-based diacylglycerol blended with palm olein, and their SFC increased with the proportion of palm olein-based diacylglycerol at all temperatures. Saberi *et al.* (2012) also showed that in making soft tub margarine, the addition of palm-based diacylglycerol to a mixture of sunflower oil and palm kernel olein increased their SFC at all temperatures.

4.3 Polymorphism

The presence of MAG and DAG can affect polymorphism in fats/oils. Most of the fats/oils composed by TAG have the form of β' crystals, but the presence of MAG and DAG stimulates the formation of β crystals. Basso *et al.* (2010) reported that the addition of MAG to palm oil induces the formation of β crystals. The addition of MAG and DAG causes the formation of crystalline nuclei in large quantities and accelerates the process of forming β crystals. Zhang *et al.* (2014) obtained that monoacylglycerol from glycerolysis of partially hydrogenated corn oil changed the type of crystal that was originally β' -crystal to β -crystal. The MAG and DAG acted as crystallization sites for the initial crystallization in the α -crystal then switched to a β -crystal due to the improved stability of the crystalline structures. Saberi, Tan and Lai (2011) found that the addition of palm-based diacylglycerol to palm oil showed that the higher the palm-based diacylglycerol was added (> 40%), the more the palm oil mixture containing high DAG was dominated by β crystals ($\beta \gg \beta'$). Saberi *et al.* (2011) also described that the crystalline form of various palm-based diacylglycerol fractions obtained the majority of crystals in the β -form. The presence of MAG and DAG acts as an accelerator in the process of crystallization of fat, then leads to the formation of β crystals. However, the presence of MAG produces higher β crystals compared to DAG (Chen *et al.*, 2014). This β crystal is very important, especially in the chocolate industry which requires fat crystals in the form of β .

4.4 Microstructure and crystalline morphology

The presence of acylglycerol (MAG and DAG) can affect the morphology of the crystal and its microstructures. Microstructures of crystals can be seen using polarized light microscopy. MAG and DAG stimulate the formation of a large number of small crystal nuclei, causing an increase in the number of crystals, and a decrease in the crystal induction period (Foubert *et al.*, 2004). Basso *et al.* (2010) reported that the addition of MAG to palm oil causes an increase in the number of crystals because MAG plays a role in

increasing the formation of crystalline nuclei that are dispersed in palm oil. Maruyama *et al.* (2014) added monoacylglycerols 1-3% to palm oil and coconut oil and showed that greater MAG addition caused smaller spherulite crystal size. Fat crystal dispersions with small size can provide desirable characteristics such as good spreadability and can inhibit the formation of grainy mouthfeel (Herrera *et al.*, 1998). Spreadability is an important characteristic of fat-based products such as butter and margarine (Rønholt *et al.*, 2013; Subroto *et al.*, 2018). Naderi *et al.* (2016) reported that the addition of MAG to the structuring of chicken fat increased the crystallized fat. The increasing MAG concentrations indicating denser structures with higher crystalline amounts of crystallized fat were seen at all temperatures (5-40°C). Likewise, Xu *et al.* (2016) reported that the morphology of crystals from palm oil and palm olein enriched with DAG showed a more homogeneous microstructure with a small form of needle-like crystals. Normah *et al.* (2014) reported that the addition of 6% DAG to palm oil caused nucleation quickly with a smaller size in palm oil.

4.5 Rheology

MAG and DAG have different rheological characteristics than TAG (fat/oil). Shakerardekani *et al.* (2013) reported that monoacylglycerol addition changed the rheological behavior of pistachio spread. The addition of 1.5% of monoacylglycerols increased the work of shear, which indicated good spreadability. Monoacylglycerols affected the rheological characteristic of pistachio spreads. Products containing monoacylglycerols showed a higher storage modulus than loss modulus. Naderi *et al.* (2018) studied the effect of monoacylglycerols on rheological properties of canola oil and reported that MAGs addition increased the viscosity, elasticity and complex moduli, and the complex viscosity. The products are suitable for the production of liquid margarine and shortenings. Phuah *et al.* (2016) studied rheological properties of palm kernel-based diacylglycerol-enriched mayonnaise, showed that rheological properties of mayonnaise displayed similar gel-like behavior with higher storage modulus and loss tangent less than 0.3. The product was suitable for healthy mayonnaise formulation. Rocha-Amador *et al.* (2014) reported that commercial saturated monoacylglycerol and mono-/diacylglycerol mixtures affected the rheological of organogels from vegetable oils. The addition of MAG and MAG-DAG mixtures (10%) increased the value of elastic modulus (G') which is associated with the solid behavior of the products. MAG and DAG organized a more stable structural configuration, due to the ease of contact between molecules, which decreases the energy needed for self-

organization and compactness of structure (Mazzanti *et al.*, 2003).

5. Utilization and application of monoacylglycerols and diacylglycerols

MAG and DAG are extensively used in the food, pharmaceutical and cosmetics industries, especially since it is known that MAG and DAG have no irritating effect on the skin compared to other ionic surfactants. MAG and DAG, specifically 1,3-DAG have positive health effects such as preventing cardiovascular disease, which makes MAG and DAG attractive produced mainly for pharmaceutical applications (Bornscheuer, 1995). MAG and DAG can be used separately or simultaneously (a mixture of MAG and DAG). MAG is the most polar compound among simple lipids. Nutritionally, MAG has similarities with TAG. MAG is completely digested and is not restricted to normal digestion (Feltes *et al.*, 2013). MAG containing oleic acid has antioxidant, anti-atherogenic, and antidiabetic properties (Cho *et al.*, 2010). This acylglycerol can inhibit the increase of TAG in the blood. MAG, both in the form of sn-1 (3) and sn-2 adjust the increase in insulin levels in the blood after digesting food to prevent obesity and diabetes due to hyperglycemia (Feltes *et al.*, 2013).

MAG is produced mainly as an emulsifier because of its better emulsification ability than DAG. Nonetheless, industrial DAG production is more attractive than MAG, especially due to its good health effects (Feltes *et al.*, 2013). DAG used as a surfactant can be applied to water in oil (W/O) emulsions such as margarine, spread, creamy filling, confectionary products, and shortening (Lo *et al.*, 2008). DAG-rich oil is also known to have beneficial physiological effects. The caloric value and absorption coefficient of DAG are almost the same as TAG. However, DAG specifically in the form of 1,3-DAG can act against cholesterol and triglyceride in the blood to inhibit obesity and prevent or reduce fat accumulation in the body (Taguchi *et al.*, 2001).

The versatility of MAG and DAG can be used or applied to a variety of fat-based products, including MAG/DAG-enriched fats/oils, shortening and margarine, surfactants/emulsifiers, and organogels as shown in Table 2.

5.1 MAG/DAG-enrich fats/oils

MAG and DAG are found naturally in fats/oils in small amounts. Nevertheless, MAG and DAG can be used to make MAG/DAG-enriched fats/oils that are safe and are even good for health (Lo *et al.*, 2008; Feltes *et al.*, 2013). Zhang *et al.* (2018) synthesized 2-MAGs rich in PUFAs by enzymatic ethanolysis of tuna oil. Under the optimal conditions, 27% of 2-monoacylglycerols was

obtained. The crystallization used acetonitrile and hexane concentrated 81.13% and 74.29% PUFA, respectively, with over 90% in the forms of 2-monoacylglycerol, which is good for health. Zhang *et al.* (2017) produced the highly unsaturated monoacylglycerol and diacylglycerol oils through enzymatic esterification and glycerolysis of oleic acid and corn oil. The product contained 46.67 wt% of MAGs and 35.56 wt% DAGs, respectively, which is suitable for palm oil-based shortening. Ferretti *et al.* (2018) synthesized diglycerides rich oil from edible vegetable oils by glycerolysis using MgO as a basic catalyst. The results showed a product containing 54% of DAG, 67% of which is the healthier 1,3-DAG. Yeoh *et al.* (2014) produced diacylglycerol enriched palm olein through glycerolysis of palm olein, which was then purified by molecular distillation. The results showed that the product has a higher slip melting point and a lower iodine number than its raw material. Saberi *et al.* (2011) synthesized palm-based diacylglycerols (P-DAG) oils by enzymatic glycerolysis of palm-based oil. The results showed that the products containing DAG of 83-90% had significantly different physicochemical properties compared to palm-based oils. Cheong *et al.* (2007) produced a diacylglycerol-enriched palm olein through enzymatic hydrolysis. The product contained 60% of DAG and 40% of TAG, which is suitable for shortening and margarine.

Modified fats/oils or structured lipids rich in MAG and DAG continue to develop along with the various functional advantages of MAG and DAG. MAG and DAG-rich structured lipids are generally synthesized via glycerolysis in one type of fat/oil (Feltes *et al.*, 2013). Synthesis of structured lipids rich in MAG and DAG by combining reactions in more than one type of fats/oils has several advantages because the SLs produced are rich in MAG and DAG, as well as having the physicochemical characteristics or desired melting profile depending on the type of fats/oils. Kowalska *et al.* (2014) successfully synthesized structured lipids rich in MAG and DAG as "fatty emulsion" from a mixture of mutton tallow and rapeseed oil through a combination of interesterification and partial hydrolysis reactions in one reaction system. Subroto *et al.* (2019) synthesized structured lipids rich in MAG and DAG through glycerolysis-interesterification of a mixture of palm stearin and palm olein. It obtained structured lipids which had better emulsion capacity and stability and higher melting point than physical blending.

5.2 Shortening and margarine

Various types of modifications to shortening and margarine have been successfully produced by making

Table 2. MAG and DAG applications in various fat-based products.

Products	Source of MAG/DAG	References
MAG-DAG Fats/Oils	Corn oil	(Zhang <i>et al.</i> , 2017)
	Vegetable oils	(Ferretti <i>et al.</i> , 2018)
	Palm olein	(Yeoh <i>et al.</i> , 2014; Cheong <i>et al.</i> , 2007)
	Palm-based oils	(Saber <i>et al.</i> , 2011)
	Mutton tallow and rapeseed oil	(Kowalska <i>et al.</i> , 2014)
	Palm stearin and palm olein	(Subroto <i>et al.</i> , 2019)
Margarine/shortening	Palm mild fraction	(Latip <i>et al.</i> , 2013)
	Sunflower oil, palm kernel olein	(Saber <i>et al.</i> , 2012)
	Canola oil	(Naderi <i>et al.</i> , 2018)
	Chicken fat	(Naderi <i>et al.</i> , 2016)
Surfactant/emulsifier	Palm based oils/fats	(Fu <i>et al.</i> , 2018)
	Palm oils	(Hattori <i>et al.</i> , 2015)
	Vegetable oil	(Maruyama <i>et al.</i> , 2014)
Organogels	Monostearate	(Ferro <i>et al.</i> , 2019)
	Monostearate and monopalmitate	(Rocha-Amador <i>et al.</i> , 2014; Lopez-Martínez <i>et al.</i> , 2015)
	Saturated fat	(Yilmaz and Ögütçü, 2014; Pérez-Martínez <i>et al.</i> , 2019)

various types of fats/oils and using acylglycerol (MAG and DAG). DAG is suitable for use in making fat products in the form of water-in-oil emulsions. Stearin fraction of palm-based diacylglycerol (PDAGS) used for the production of bakery shortening obtained that a mixture of PDAGS and palm mild fraction (PMF) produced bakery shortening with characteristics of better structural compatibility and full miscibility with mixed crystals $\beta + \beta'$ polymorphs (Latip *et al.*, 2013). The saturation of fatty acids in MAG and DAG also affects the storage stability of the resulting shortening. MAG and DAG which are rich in unsaturated fatty acids are known to decrease the crystallization rate, improved stability of β' crystals, and produce a more stable hardness (Zhang *et al.*, 2017). DAG can also be used for making margarine. Saber *et al.* (2012) produced palm-based diacylglycerol-enriched soft tube margarine from a mixture of palm-based diacylglycerol, sunflower oil, and palm kernel olein with a ratio of 50/35/15. It obtained margarine with a similar SFC profile, but saturated fatty acids are lower compared to commercial margarine. Naderi *et al.* (2018) utilizing MAG in making liquid margarine and shortening from canola oil obtained products suitable for use in bakery. MAG from palm oil can also be mixed with chicken fat to get structured chicken fats that can be applied to produce soft tub margarine and Iranian vanaspati (Naderi *et al.*, 2016)

5.3 Surfactant/emulsifier

MAG and DAG are compounds containing hydrophilic and hydrophobic groups making them emulsifiers that can form stable and homogenous emulsions (Devi *et al.*, 2008). MAG and DAG supply about 75% of the world's emulsifier production for food (Feltes *et al.*, 2013). MAG is widely used in bakery,

margarine, milk and confectionery products, especially as an emulsifying agent and stabilizer (Damstrup *et al.*, 2005). For applications as food emulsifiers, MAG should have a high purity because MAG has better emulsification properties compared to mixtures of different acylglycerols (Bornscheuer, 1995). MAG is often used as an emulsifier, where the use of 0.5% by weight of the material is usually obtained by a stable emulsion (Feltes *et al.*, 2013). Fu *et al.* (2018) using MAG and DAG as emulsifiers in shortening for cake making, found that MAG and DAG encourage inclusion and retention of air in cake dough, thereby increasing the volume and structure of cake crumbs. Emulsifiers, in general, can improve the hardness, compactness, suppleness, and the level of staling in cakes (Paraskevopoulou *et al.*, 2015). MAG, as an emulsifier also has the ability to form complexes with starch, which can improve the quality of bread, such as firmness and hardness (Hattori *et al.*, 2015). MAG and DAG act as emulsifiers, which can inhibit fat bloom, as it is known that emulsifiers can prevent susceptibility to fat bloom that often occurs in chocolate products (Dhonsi and Stapley, 2006; Smith *et al.*, 2011).

5.4 Organogels

Lipid organogels are an alternative to getting semi-solid fat by reducing the proportion of saturated fatty acids, making them safer for health (Chaves *et al.*, 2018). MAG and DAG can improve organogel microstructure of vegetable oil with better intermolecular forces (Rocha-Amador *et al.*, 2014). The use of MAG in the form of glyceryl monostearate has been proven to be able to improve the crystals and texture of oleogels (Ferro *et al.*, 2019). Lopez-Martínez *et al.* (2015) developed MAG-organogel with and without ethylcellulose from

vegetable oil. The results showed that there was a synergistic interaction between MAG and ethylcellulose, which resulted in organogels with lower oil loss and better viscoelastic properties than without ethylcellulose. Yilmaz and Ögütçü (2014) developed hazelnut oil organogels with beeswax and MAG as organogelators. It was found that the higher the organogelators, the faster the formation of crystals and SFC increased. The organogels obtained are also more resistant to oxidation and can be used as a basis for margarine and shortening. Pérez-Martínez *et al.* (2019) also showed that MAG can act as nucleation sites but does not affect the thermodynamic stability of candelilla wax oleogels. Thus, MAG and DAG have the potential to improve oleogel and organogel products

6. Conclusion

MAG and DAG can improve various physicochemical properties of fats/oils for various purposes that require higher melting and crystallization temperatures, fat crystals in the form of β , smaller and homogeneous crystallite sizes, and harder textures or higher viscosities. MAG and DAG have been widely applied in various fat-based products and improve the characteristics of products such as MAG/DAG-enriched fats/oils, margarine/shortening, surfactants, and organogels.

Conflict of Interest

The author declares no conflict of interest.

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