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The Effect of monoglycerides on the crystallisation of triglyceride

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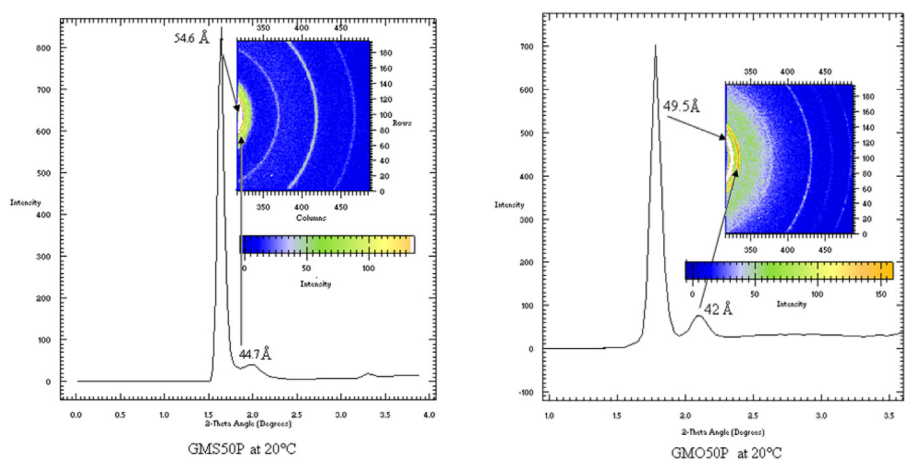
HIGHLIGHTS

- The effect of monoglycerides on the crystallisation of oils was investigated.
- The polymorphism of miglyol oil and palm oil changed with the presence of GMS.
- The polymorphism of olive oil and palm oil were not affected by the presence of GMO.

GRAPHICAL ABSTRACT



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ABSTRACT

There has been increased interest recently in how different lipids interact with each other in solid phases and in the properties and consequences of the inter-lipid associations. The crystallisation behaviour of the triglycerides is noticeably influenced by the presence of monoglycerides. The main aim of this work was to extend studies of the influences of saturated and unsaturated monoglycerides on the crystallization of triglycerides. We aim to gain more insight into the effect of monoglycerides on miglyol oil, palm oil and olive oil crystallization. Optical microscopy, differential scanning calorimetry (DSC), small X-ray scattering (SAXS) and wide X-ray scattering (WAXS) were used in this study. Our results suggest that the presence of saturated monoglyceride affects the oil crystallization behaviour. Conversely, triglycerides with unsaturated fatty acid chains show little impact. The results provide evidence that saturated and unsaturated monglycerides are incorporated into the α- phase of palm oil.

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1. Introduction

Monoglycerides are important food emulsifiers and are used in personal care products as well as for drug delivery [1–3]. There has been an increased interest recently in how different lipids

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Table 1
Nomenclature of lipid phases.

Nomenclature	Phase name	Packing modes
L_2	Isotropic phase	Disordered
α-form	α Phase for diglyceride and triglyceride	Hexagonal lattice
Lβ	α-gel phase	Hexagonal lattice
$L_{\beta 1}$	New structure of α- gel phase	Hexagonal lattice
Sub α phase	Sub α phase	Orthorhombic lattice
β‘	β′-crystal phase	Orthorhombic lattice
β_1 crystal	Glycerol monooleate GMO crystallization phase	Triclinic lattice
β_2 crystal	Oil crystallization phase	Triclinic lattice
β_3 crystal	Glycerol monostearate GMS crystallization phase	Triclinic lattice

interact with each other and in the properties and consequences of the inter-lipid associations [4–8]. The crystallisation behaviour of the triglycerides is noticeably influenced by the presence of polar lipids with saturated fatty acid chains. Conversely, lipids with unsaturated fatty acid chains show trivial impact [9]. Small angle X-ray scattering SAXS data revealed that crystallisation of triacylglycerols for the mixture of miglyol oil and cocoa butter successively occurred with α-form 2L (49.3 Å) structure, then β′ form (Orthorhombic lattice) exhibited peak at 44.5 Å and 34.5 Å [9]. Monoacylglycerols behave as nucleation seeds for palm oil thereby reducing the crystallization time [10]. Furthermore, addition of monoacylglycerols does not change the final solids content after isothermal crystallisation at 25 °C. These findings are concordant with what was demonstrated by Wright et al. [11]. Niiya et al. [12] revealed that addition of saturated monoglycerides to palm kernel oil both increased the melting point and induced the crystallisation of palm kernel oil along with increasing the solid fat content. By contrast, addition of unsaturated monoglycerides decreased the melting point of palm kernel oil, but accelerated palm kernel crystal growth [12].

The addition of lauric acid derivatives increase the crystal growth rate of trilaurin [13]. Fourbert et al. demonstrated that the presence of partial glycerides influences the crystal growth and nucleation of triglycerides, the interaction between partial glycerides and triglycerides depends on the number of fatty acids esterified and the concentration of the partial glycerides. Monostearien improves nucleation at low temperature and effects the crystal growth at higher temperature, while monoolein influences the nucleation process but has no effect on crystal growth [14]. These finding are in agreement with the results observed by Basso et al. [10]. Fredrick et al. [15] studied the effect of monoglycerides of hydrogenated palm oil on the palm oil crystallisation behaviour as a function of time using X-ray scattering studies. An initial peak in the SAXS diffraction pattern at 52.5 Å indictaed the presence of α-phase monoglyceride, a second peak appeared at 46.5 Å, which corresponded to palm oil. The latter peak vanished and a third peak appeared at 42.19 Å. The formation of α-phase was also observed from a WAXS peak at 4.15 Å. β′phase appeared at two peaks 4.2 Å and 3.8 Å [15]. Monoglycerides of hydrogenated palm oil accelerate the process of palm oil crystallisation whereas monoglycerides of hydrogenated sunflower oil had no effect on the crystallisation of palm oil [15]. The difference was attributed to the homogeneity of the fatty acid chains of palm oil and the monoglycerides of hydrogenated palm oil versus the lack of homogeneity in the case of monoglycerides of hydrogenated sunflower oil [15] (Table 1).

Basso et al. [10] explained the mechanism of how the presence of monoacylglycerol induces palm oil crystallisation. Monoacylglycerols behave as nucleation seeds when dispersed within the palm oil. Subsequently, palm oil triglycerides undergo adsorption on the seed surface. Palm oil polymorph changes from β′ to β-crystals by the addition of monoacylglycerols [10]. The addition of polyglycerol behenic acid to palm oil induces the formation of β′ phase without transformation into β-crystal during the heating process.

Table 2
The alkyl chain distribution of miglyol 812 oil, palm oil and olive oil.

Material	C6	C8	C10	C12	C14	C16	C18:0	C18:1	C18:2
Miglyol oil	2%	50%	45%	2%	1%				
Palm oil					1%	44%	39%	10%	6%
Olive oil						13%	3%	77%	7%

However, in the absence of the additives, palm oil crystallizes into the α-form after instant cooling. The α-form, when heated, converts to the β′-form which in turn transforms to the β-form [16]. Miglyol oil (M) is a medium saturated alkyl chain triglyceride that is widely used in food manufacturing, nutritional supplements and for drug delivery [17–19]. Palm oil (P) is frequently used in food processing due to the formation of β′ phase required for the smooth texture in food products [20]. Palm oil contains more long saturated fatty acids of triglycerides compared to all other vegetable oils [21]. Olive oil (O) represents one of the most commonly used edible vegetable oils and is becoming a widespread product due to evidence of its beneficial health effects [22]. Olive oil is considerably rich in monounsaturated alkyl chains of triglycerides. Great attention is paid to foods and drugs with unique sensorial performance; some authors have investigated the use of monoglyceride self-assembly to control the flavour release in low fat foods [23]. The main aim of this work was to extend studies on the influences of saturated and unsaturated monoglyceride on the crystallization of triglycerides to elucidate the mechanism of oil crystallization and gain more insight into the effect of monoglycerides on miglyol oil, palm oil and olive oil crystallization behaviour. Optical microscopy, differential scanning calorimetry (DSC), small X-ray scattering (SAXS) and wide X-ray scattering (WAXS) were used in this study. Our results suggest the presence of saturated monogleceride affects the oil crystallization behaviour.

2. Experimental

2.1. Materials, sample preparation

Glycerol monooleate (GMO) and glycerol monostearate (GMS) were obtained from Givaudan. The GMO and GMS formulations contain greater than 90% monoglycerides and less than 10% diglyceride and triglyceride [24]. The alkyl chain distribution of miglyol812, palm oil and olive oil are given in Table 2.

Weighed mixtures of monoglycerides and oils were heated to 80 °C and held constant for 30 min using a hot plate with magnetic stirrer; different samples of the monoglycerides with varying oil content were examined. The abbreviations for each of the samples are listed in Table 3.

2.2. Scanning electron microscopy

The instrument used was an FEI Quanta 200 Environmental SEM, which has a high resolution of 3.0 nm. The instrument chamber

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