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Effect of Fatty Acid Composition of Monoglycerides and Shear on the Polymorph Behavior in Water-in-Palm Oil-Based Blend

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Abstract Effect of the fatty acid composition of mono-glycerides and shear on the polymorphic behavior in a water-in-oil (W/O) emulsion semi-solid fats blend was investigated. The bulk fat blend and the W/O emulsion fat blend with added either unsaturated or saturated mono-glycerides were prepared using a rapid cooling heat exchanger, and the polymorphic transitions from β' to β-form during storage were compared by X-ray diffraction. The peak intensity of the β-polymorph of the samples with added saturated monoglycerides was stronger than that of unsaturated monoglycerides, and the peak intensity of the β-polymorph of the W/O fat blend emulsions was stronger than that of the bulk fat blends. The polymorphic transition to the β-form during storage of the W/O emulsion fat blend was promoted by the applied shear. In contrast, that of the bulk fat blend was retarded by shear. These results sug-gested that fatty acids, which combined to monoglycerides on the surface of water droplets, influenced the polymorph transformation of the fat crystal network in a continuous semi-solid fat phase because of the acyl–acyl interactions between the fatty acid residues of the monoglycerides and triacylglycerols in the fat crystal network.

Keywords Fats and oils · Fat crystallization · Lipid chemistry · Lipid analysis Polymorphism · Process control · Processing technology

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Introduction

Margarine and fat spreads are water-in-oil (W/O) emul-sions consisting of semi-solid fats, water, emulsifiers, and other ingredients. The semi-solid fats include fats with high melting points that form crystal network structures and provide better physical properties for these products. Palm oil is one of the major fat sources in the world because of its heat stability, physical characteristics, availability, price competitiveness, and no *trans* fatty acids. However, when palm oil is used in margarine and fat spread, granular crystals often crystallize, causing margarine to deteriorate. Many studies have been conducted to clarify the formation mechanism of these granular crystals in palm oil-based margarine [1–3]. Tanaka et al. [4] examined the effect of higher melting fat such as tripalmitoylglycerol (PPP) on the formation of granular crystals and concluded that PPP promoted the formation of granular crystals in margarine. Humphrey and Narine [5] highlighted the importance of the phase behavior of PSP, PPS, PSS, and SSS (S: stearic acid, P: palmitic acid) to typical industrial shortening systems. Watanabe et al. [6] found that the crystallization of granular crystals in palm oil-based margarine was closely related to the polymorphic transition from the β' to the β-form of 1,3-dipalmitoyl 2-oleoyl glycerol (POP). Miura et al. [7] determined that the POP content of the granular crystals in palm oil-based margarine was higher than that in the surrounding materials. Tanaka et al. [8] suggested that the crystallization and transformation of tripalmitin and tristearin fractions using microbeam X-ray diffraction (XRD) promoted the microstructures and for-mation processes of granular crystals in relation to the fractional crystallization of the β-form of POP.

Table spreads are multiphase colloidal systems con-sisting of an aqueous phase dispersed as droplets within a

continuous oil phase and a fat crystal network. The fat crystal network prevents the destabilization of water droplets from coalescing with the crystallized emulsifiers that are adsorbed at the oil–water interface [9, 10]. Rousseau et al. [11] found that the concerted role of both the solid fat crystal network and interfacial fat crystals dictated the dispersed phase stability. The crystals interact with each other and form a three-dimensional network, providing these spreads a hard, solid character. Surfactants are added to fat products to increase the stability of the dispersed phase. The margarine manufacturing industry widely uses monoglycerides in W/O emulsion systems to increase their stabilities. Some emulsifiers, such as sucrose and sorbitan fatty acid esters, can be used to control fat crystallization properties such as crystal morphology [12]. Wassell et al. [13] reviewed some techniques to control the trans and saturated fatty acid content using crystal structure modifiers and co-gelators. Macierzanka et al. [14] reported the effects of the crystalline emulsifier composition on the structural transformations of W/O emulsions. Although some studies have been performed on the crystallization properties of emulsifiers in emulsion systems, information about the relationship between the structures of monoglycerides added to W/O emulsions and polymorph behavior during storage conditions is still unclear.

In the food industry, fat crystals in semi-solid food products are formed during the cooling process [15]. The bulk crystallization process is sensitive to parameters such as temperature, shear forces, and the fat phase composition. Humphrey and Narine [5] compared the lipid shortening functionality as a function of the molecular ensemble and shear. Miskandar et al. [16] have studied the processing conditions, such as cooling treatment and pin working which influence the physical properties of palm oil based margarines. Many studies have reported the effects of shear applied during the production process and the microstructures of fat products on the polymorph structures [17–21]. However, the effects of shear on the polymorph behavior of W/O emulsions during storage have been investigated very little.

A major objective of this study was to investigate the emulsifiers and the processing conditions on the crystallization of a W/O emulsion palm oil-based semi-solid blend. In this study, we showed the impact of the fatty acid composition of monoglycerides and shear on the polymorph behavior of W/O emulsions during storage.

Materials and Methods

Preparation of Fat Blends

Table 1 shows the major ingredients used in the production of the bulk fat blend (100% fat) and the W/O emulsion fat

Table 1 Formulations of the fat blend samples

%	Bulk fat blends	W/O emulsion fat blends
Fully hydrogenated rapeseed oil	2.84 (2.0[a])	2.0
Palm oil	28.41 (20.0[a])	20.0
Soybean oil	68.19 (48.0[a])	48.0
Monoglycerides[b]	0.5	0.5
β-Carotene	0.001	0.001
Water	0.0	29.4
Total	100.0	100.0

[a] Ratio in total oil phase

[b] Oleate, palmitate, stearate, and behenate

Table 2 Fatty acid compositions of total fats

Fatty acid	%
C12:0	0.1
C14:0	0.3
C16:0	19.5
C18:0	5.3
C18:1 cis	26.7
C18:2 cis	38.9
C18:2 trans	0.2
C18:3 cis	4.7
C18:3 trans	0.7
C20:0	0.6
C20:1 cis	0.2
C22:0	1.6
Others	1.2
Total	100

blend (70% fat). The palm oil-based fat blend was prepared with soybean oil, palm oil, and fully hydrogenated rapeseed oil. The *trans* fatty acid content was <1.0%. This fat blend's polymorph had a tendency to transform to the β-form [7, 8]. The main saturated fatty acids of the fat blends were palmitic acid (C16:0) and stearic acid (C18:0) (Table 2). A constant amount (0.5 wt%) of monoglycerides (either oleate, palmitate, stearate, or behenate) was also added. Riken Vitamin Co. Ltd. (Tokyo, Japan) supplied the distilled monoglycerides. Table 3 lists the fatty acid compositions of the monoglycerides. We prepared the fat blends following Hui's procedure [1]. All the oils were melted and mixed at 60 °C, followed by adding the emulsifiers with gentle agitation. We added the aqueous phase to the oil phase with agitation at 50 rpm for 10 min to form a W/O emulsion. The emulsion was cooled using a rapid cooling heat exchanger (Schröder GMBH & Co., Germany) to form a semi-solid fat structure. The temperature of the fat mixtures at the inlet and outlet of the heat

Table 3 Fatty acid compositions of the monoglycerides used in this study

%	Oleic acid MG	Palmitic acid MG	Stearic acid MG	Behenic acid MG
C12:0	0.0	0.3	0.0	0.6
C14:0	0.0	1.1	0.1	0.0
C16:0	3.8	43.6	10.8	4.6
C18:0	3.1	53.8	86.3	9.9
C18:1c	85.3	0.0	0.1	0.0
C18:1t	0.3	0.4	0.8	0.2
C18:2c	5.4	0.0	0.1	0.2
C20:0	0.3	0.5	0.7	4.1
C22:0	0.9	0.1	0.7	79.1
Others	0.9	0.1	0.3	1.3
SFA	8.1	99.4	99.0	99.6
Total	100.0	100.0	100.0	100.0

SFA Saturated fatty acids

exchanger were 60 and 10 °C, respectively. The final product was poured into 160-g tubs as the samples without shear. The samples with applied shear were obtained by passing the product through a pin machine. The shaft rotating speed was maintained at 130 rpm; the temperatures at the product inlet and outlet were 2.3 and 6.4 °C, respectively. The samples obtained were kept at 5 °C for 3 days. The samples were stored using two different storage conditions: one set was subjected to thermal thawing between 5 and 20 °C every 12 h, while the other was continuously stored at 5 °C.

XRD Measurements

We carried out XRD measurements (Rint-Ultima 2000, CuK α:$\lambda = 1.54$ Å; Rigaku; Tokyo, Japan) to determine the wide-angle diffraction patterns ($2\theta = 18°–25°$) and to calculate the short spacing of the crystal polymorphs. The sample was maintained at 5 °C during these measurements to prevent the crystals from melting. We performed duplicate analyses of these measurements, and the reproducibility of all the data was confirmed for all measurements.

Solid Fat Content

The solid fat content (SFC) was determined with an NMR Analyzer mq20 (Bruker Optik GmbH; Ettlingen, Germany). Samples were obtained by inserting plastic cylinders into the spread at random locations by hand. The filled cores were then transferred directly into NMR tubes (ID = 0.8 cm, L = 18 cm). Triplicate analyses were performed on all SFC measurements.

Droplet Size Distributions

We used electron microscopy to examine the droplet distributions of the W/O emulsion fat blends. Samples were observed using a cryo-scanning electron microscope S-4300 (Hitachi High-Technologies Corp., Tokyo, Japan). An accelerating voltage of 2 kV was used with a magnification of 3,000. Numerous images of a typical field of each sample were captured. Image analysis software (A-zoukun; Asahi Kasei Engineering Co., Tokyo, Japan) was used to calculate the droplet diameter from four representative images of each sample.

Polarized Light Microscopy

About 50 mg of a sample was placed on a cooled glass plate followed and covered with a cover glass. The cover glass was manipulated using a wooden-needle until the sample layer was thin enough to observe the crystals. A Microphot-FX polarization microscope (Nikon, Tokyo, Japan) was used.

Statistical Analyses

We performed ANOVA and *t* tests to determine the relative proportion of β-polymorph, SFC, and droplet size. Statistical differences were considered significant at $P = 0.05$.

Results and Discussion

The Differences of the Polymorphic Structure Between the Bulk Oil and the W/O Emulsion

Figure 1 shows the wide-angle XRD profiles of the bulk fat blend and W/O fat blend emulsion, either with added 0.5 wt% oleic acid or palmitic acid monoglyceride, after 3 days of sample preparation. We characterized the wide-angle XRD peak at 0.46 nm as the β-form and those at 0.42 nm and 0.38 nm as the β'-form. The XRD profiles of the bulk fat sample with oleic acid monoglyceride (a) corresponded to the typical structure of margarine fats with the β'-form. In contrast, the W/O fat emulsion sample with oleic acid monoglyceride (b) had a weak peak at 0.46 nm, characterized as the β-form. The XRD profile of the bulk fat sample with palmitic acid monoglyceride (c) had a weak peak at 0.46 nm. The intensity of the peak at 0.46 nm of the W/O fat emulsion sample with palmitic acid monoglyceride (d) was greater than that of the bulk fat blend. Figure 2 shows the relative β-form intensities of the bulk fat blend and W/O fat emulsion blend after 3 days of sample preparation. The y-axis is the relative proportion of the β-polymorph (R β), calculated as the ratio of the peak

height of the 0.46 nm line over the height of the sum of the peak heights 0.38, 0.42, and 0.46 nm. The R β of the W/O emulsion fat blend with added oleic acid monoglyceride was significantly greater than that of the bulk fat blend with added oleic acid monoglyceride ($P < 0.05$), and the R β of the bulk fat blend with added palmitic acid monoglyceride was greater than that of the bulk fat blend with added oleic acid monoglyceride ($P < 0.05$). The W/O emulsion fat blends, containing approximately a 30% water phase, existed as dispersed water droplets in a continuous semi-solid fat. These results indicated that the existence of the dispersed aqueous droplet phase and the fatty acid composition of monoglycerides influenced the polymorphic structures. The saturated fatty acids residues in oleic acid monoglyceride (8.1%) and palmitic acid monoglyceride (99.4%) on the surfaces of the water droplets likely interacted with fatty acids in the triglycerides such as POP and acted as templates for the crystallization and polymorphic transition in the fat network.

Effect of Thermal Thawing

Figure 3 shows the effects of thermal thawing between 5 and 20 °C (temperature range in which fat spreads are often consumed) every 12 h on the phase transition behavior of the bulk fat blend and W/O emulsion fat blend prepared with oleic acid monoglyceride. The R β of the samples stored at 5 °C increased very slowly during 28 days. In contrast, the R β of the samples with thermal thawing increased remarkably with increasing storage periods up to 28 days. Table 4 shows the SFC of these samples at 5 °C and 20 °C. The SFC in the fat blend with added oleic acid monoglyceride decreased from 17.6 to 7.1% with increasing temperature from 5 °C to 20 °C. This drop was due to the melting of low and medium-melting triglycerides such as POO (melting point 18.0–19.0 °C). Johansson et al. [19] reported that the strength of the fat crystal networks increased at elevated temperatures on account of the increased adhesion between the partially melted crystal surfaces. Garbolino et al. [12] reported that the high temperature during cycling induced the polymorphic transition of POP. The effects obtained from thermal thawing were in accordance with these previous reports. These results showed that thermal thawing provided the driving force for the polymorphic transition from the β' to the β-form, including the melting process of the medium-melting triglycerides.

Fig. 1 X-ray diffraction of fat blend samples. **a** Bulk fat blend with added oleic acid MG, **b** W/O emulsion fat blend with added oleic acid MG, **c** bulk fat blend with added palmitic acid MG, and **d** W/O emulsion fat blend with added MG
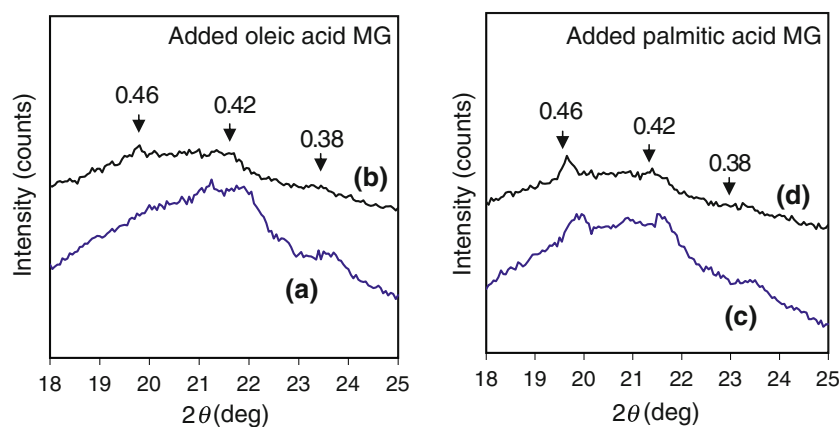

Fig. 2 Relative β intensity (R β) of the X-ray diffraction profiles of fat blend samples. **a** Bulk fat blend and **b** W/O emulsion fat blend
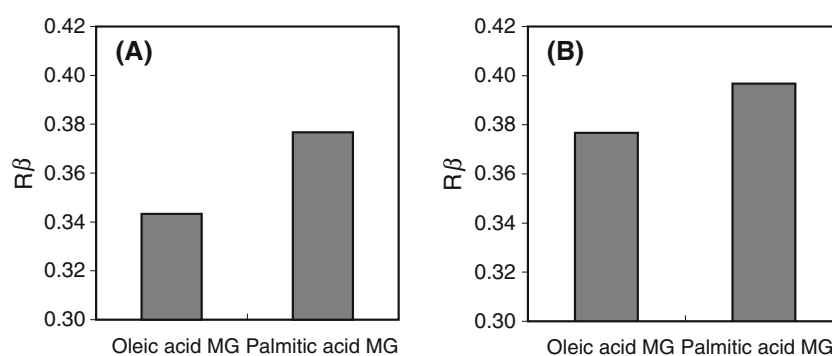
Fig. 3 Effects of thermal thawing on the polymorph transition of the bulk fat blend and the W/O emulsion fat blend with oleic acid monoglyceride. **a** Bulk fat blend kept at 5 °C, **b** bulk fat blend with thermal thawing between 5 and 20 °C every 12 h, **c** W/O emulsion fat blend kept at 5 °C, and **d** W/O emulsion fat blend with thermal thawing between 5 and 20 °C every 12 h
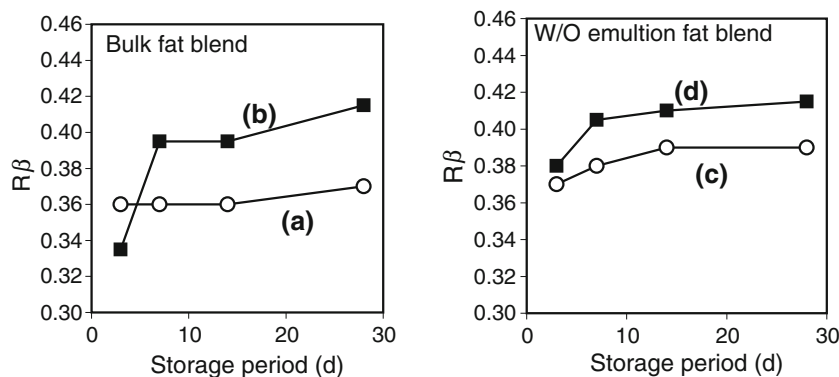

Table 4 SFCs of the fat blends

%	5 °C	20 °C
Bulk fat added oleic acid MG	17.6 ± 0.2 (12.3^a)	7.1 ± 0.1 (5.0^a)
Bulk fat added palmitic acid MG	18.5 ± 0.3 (13.0^a)	7.2 ± 0.2 (5.0^a)
W/O fat added oleic acid MG	14.5 ± 0.3	5.5 ± 0.2
W/O fat added palmitic acid MG	14.4 ± 0.2	6.0 ± 0.4

[a] 70% of SFC

Effects of the Fatty Acids of Monoglycerides on the Polymorph Behavior during Storage

We evaluated the time dependence for the polymorphic transformation of the bulk fat blend and the W/O emulsion fat blend with either added oleic acid or palmitic acid monoglyceride under thermal thawing conditions. The R β of the four samples increased with an increasing storage period (Fig. 4). The bulk fat blend with added oleic acid monoglyceride displayed the lowest R β of the four samples tested 3 days after the preparation. In contrast, the R β of this sample reached the same level of the other samples after 28 days. We observed more pronounced differences in the R β values during storage for the bulk fat blend with added oleic acid monoglyceride. The phase transition to the β-form of the samples with added palmitic acid monoglyceride was faster than that of the sample with added oleic acid monoglyceride for the bulk fat and the W/O emulsion fat samples. These results showed that the fatty acid compositions of monoglycerides affected the polymorph behavior during storage.

Effects of Shear on Polymorph Transformation

We evaluated the effects of shear on the polymorph behavior of these samples during storage. Figure 5 shows the polymorphic transformation of the bulk fat blend and the W/O emulsion fat blend with and without shear under thermal thawing. The R β of the bulk fat sample 3 days
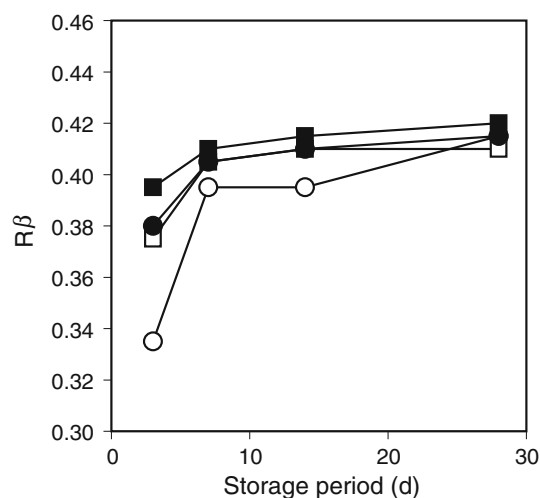

Fig. 4 Changes of the relative β intensity (R β) of the X-ray diffraction profiles of the fat blend samples: bulk fat blend with added oleic acid MG (*open circles*), W/O emulsion fat blend with added oleic acid MG (*filled circles*), bulk fat blend with added palmitic acid MG (*open squares*), and W/O emulsion blend with added palmitic acid MG (*filled squares*)

after preparation without shear was less than that of the sample with shear. However, the R β of the bulk fat sample after 28 days without shear was greater than that of the sample with shear. Whereas the R β of the W/O emulsion fat blends without shear were less than those of the fat blends with shear both after 3 and 28 days. These results showed that shear on the bulk fat blend retarded the polymorphic transition from the β' to the β-form, while shear on the W/O emulsion fat blend did not significantly affect the polymorphic transition during storage. Table 4 showed that 70% of the SFC of the bulk fat blends was less than that of the W/O emulsion fat blends. And bulk fat blends have relatively larger free space for partially melting of solid fats and subsequent re-construction of fat network by thermal thawing. These results suggested that the triglycerides related to the phase transition in the bulk fat were more easily transferred than those in the W/O emulsion fat blends [8, 20]. Crystals interacted with each

Fig. 5 Polymorph transition of the two systems of the fat blend with and without shear: **a** bulk fat blend with added oleic acid MG, **b** bulk fat blend with added palmitic acid MG, **c** W/O emulsion fat blend with added oleic acid MG, and **d** W/O emulsion fat blend with added palmitic acid MG
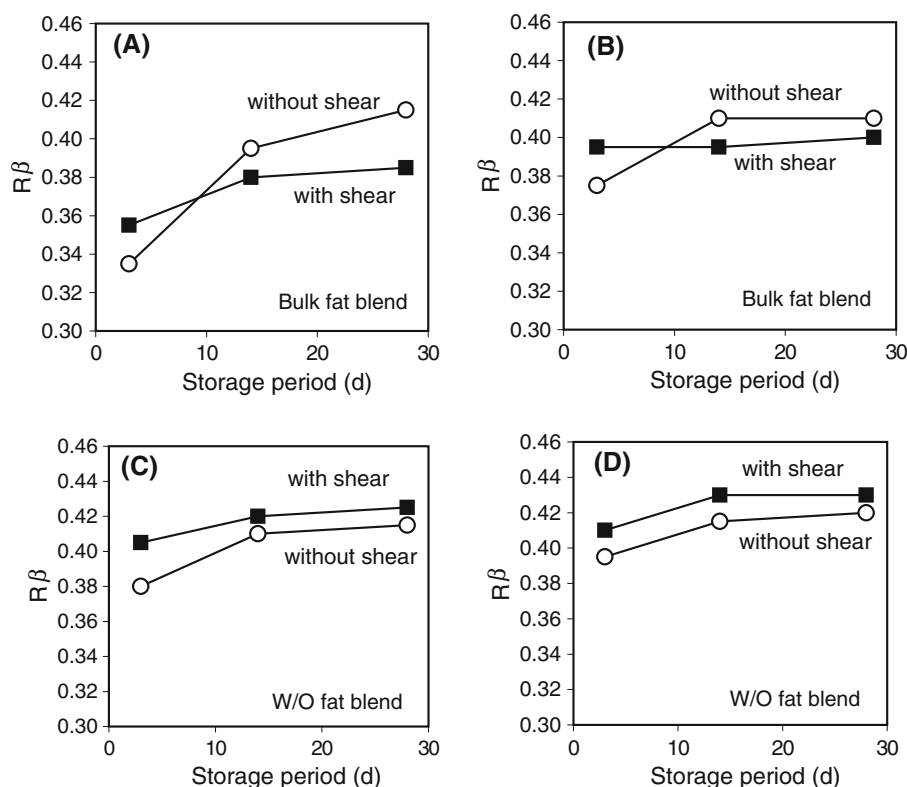
other and formed a three-dimensional network that gave these spreads a hard, solid character [21]. The interactions between the crystals were influenced by the adsorption of food emulsifiers. Fat crystals in the W/O emulsion fat blends were present in both the surface-active crystals on the interface and the networked interstitial crystals between the water droplets [9, 18]. The surface-active crystals at the interface exposed the hydrophobic surface towards the oil and the hydrophilic surface towards the water. The greater heat stability of the surface-active crystals likely stabilized the fat crystal network against the polymorphic transformation induced by thermal thawing because the polymorphic transition from the β' to the β-form in the bulk fat blend was faster than that of the W/O emulsion fat blends. The R β of both the bulk and the W/O emulsion fat blends with shear after 3 days were greater than those of the blends without shear, and the changes in these R β values with shear during storage were relatively small. Shear strongly influenced the crystal network structure that formed and accelerated the solid-state phase transformation [17, 22]. Mazzanti et al. [20] demonstrated using synchrotron XRD that shear-induced crystalline orientation and phase transition acceleration occur in different edible fats, cocoa butter, milk fat, and palm oil. Our results were in agreement with these previous reports. This study demonstrated the shear-induced acceleration from less stable to more stable crystalline polymorphic forms in pin-working margarine processing conditions.

Effects of Aqueous Droplets and Crystals on Polymorph Behavior

To evaluate the influence of the distribution of water droplets on polymorph behavior, we determined the droplet size of the W/O emulsion. The average sizes of the droplet diameter of the W/O emulsion fat blends were calculated from the electron microscope images. The average droplet sizes obtained for the samples with oleic acid monoglyceride with and without shear were 1.18 and 1.24 μm, while those obtained for the sample with palmitic acid monoglyceride with and without shear were 1.46 and 1.45 μm, respectively. Figure 6 shows the droplet distribution of the samples with added oleic acid monoglyceride with and without shear. These samples had narrow droplet size distributions, and there were no significant differences for the droplet size distributions between samples with and without shear ($P > 0.05$). This result indicated that the shear applied in this study did not influence the droplet sizes. Figure 7 shows the polarized light microscopy (PLM) photomicrographs of the bulk fat blends and the W/O emulsion fat blends with added oleic acid monoglyceride. Dark regions of the W/O emulsion fat blends in the PLM images are aqueous droplets. The crystals were much smaller in size than the droplets. We did not observe any differences in the PLM images between the samples with and without shear. These results indicated that the differences in the polymorph behavior with and without

shear were not due to the differences in the aqueous droplet and fat crystal sizes.

Effects of Fatty Acid Compositions of Monoglycerides

We compared the R β of the W/O emulsion fat blends prepared with three saturated fatty acid monoglycerides, palmitic, stearic, and behenic acid, to investigate the influence of the saturated fatty acid composition of the monoglycerides on the polymorphic structure (Fig. 8). Although the R β of the samples with added palmitic acid monoglyceride was greater than that of oleic acid

Fig. 6 Distributions of the aqueous droplet diameters with and without shear

($P < 0.05$), no significant difference was observed among the saturated fatty acid monoglycerides tested ($P > 0.05$). Monoglycerides added to the W/O emulsions existed on the interface of the aqueous droplets. Saturated fatty acid monoglycerides probably influenced the polymorph structures of the surrounding crystals on the aqueous droplets and affected the subsequent polymorph transformations of the networked interstitial crystals between the droplets. If the acyl groups of the emulsifiers and triacylglycerols were similar, their interaction was enhanced [11]. Palmitic and stearic acid monoglycerides occupied approximately 95% of the total saturated fatty acids in the fat blends (Table 2). The saturated acyl chains of the monoglycerides probably interacted with the acyl chains of the major saturated fatty acids in the triacylglycerols of the fat blends, such as PPP, SSS, and POP, and could induce the β-form polymorph of POP. It was reported that addition of monopalmitoylglycerol and monostearoylglycerol to palm oil promoted the solidification of the model cream while the addition of mono-behenoylglycerol had no such effect [23]. This report suggested that the combination of specific fatty acid species on monoglycerides with palm oil may be a key to this crystallization behavior. A more detailed investigation is necessary to understand the interrelation of the fatty acids of monoglycerides and triglycerides in W/O semisolid emulsions. These results showed that the selection of the fatty acid composition of monoglycerides was essential for controlling the polymorph behavior of the fat blends containing palm oil.
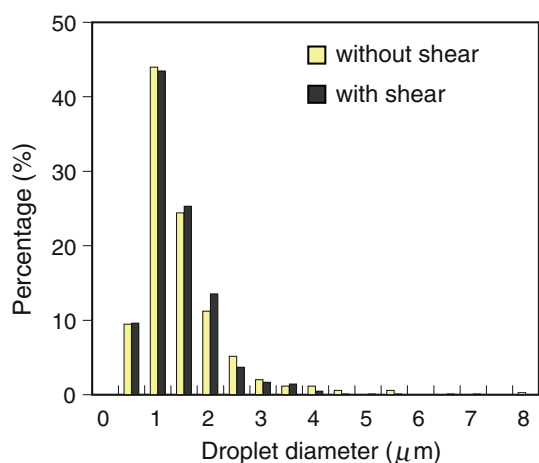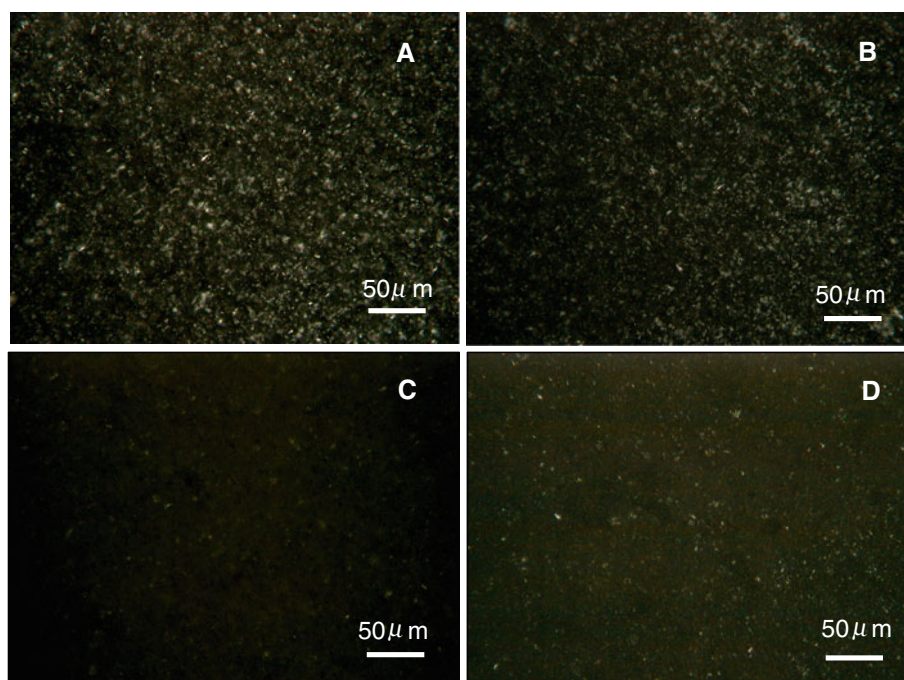
Fig. 7 Polarizing microscope images of fat crystals. **a** Bulk fat blend with added oleic acid MG with shear, **b** bulk fat blend with added oleic acid MG without shear, **c** W/O emulsion fat blend with added oleic acid MG with shear, and **d** W/O emulsion fat blend with added oleic acid MG without shear
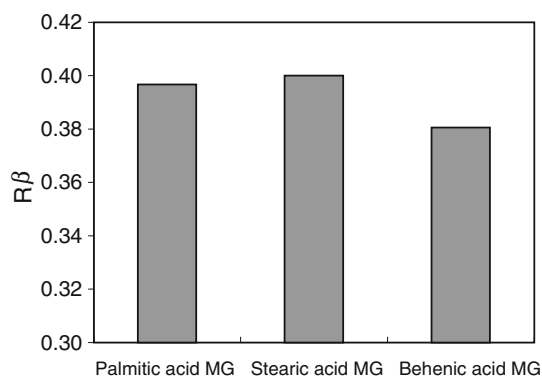
Fig. 8 The relative β intensity (R β) of the X-ray diffraction profiles of the fat blend samples with added saturated fatty acid monoglycerides

Generation of Granular Crystals

Among the samples used in this study, we observed granular crystals with the naked eye only in the bulk fat blend with added oleic acid monoglyceride without shear under the thermal thawing conditions. Figure 9 shows the granular crystal (approximately 3 mm diameter) that appeared in this sample. Fat blends containing palm oil have been reported to include tendency for polymorphic transition to β-form [2, 3, 16], and granular crystals in the palm oil blend likely related to the fractional crystallization of the β-form of POP [8]. Despite the fact that all samples tested in this study showed the β-polymorph after 28 days, granular crystals were observed only in the sample of the bulk fat blend with added oleic acid monoglyceride without shear. The distinguishing characteristic of this bulk sample with oleic acid monoglyceride are included below. The initial R β was relatively low, resulting from the lack of inducing effect for the generation of crystals in the fat blend by unsaturated fatty acid monoglycerides, and the shear did not add any promotional effects on the polymorphic transformations. Significant differences in the SFC of the bulk fat blends between 5 and 20 °C by thermal thawing might allow the possibility of fat crystals in the polymorphic transformation. In addition, all fat crystals in the bulk fat existed as network crystals as against the W/O emulsion fat blends in which the solid fat was constructed with interfacial crystals around aqueous droplets and network crystals [14]. These characteristics resulted in the largest degree of changes in the polymorphic transformation, and the slow rate to the β-form during storage among the fat blends was investigated. We concluded that the rate of the phase transition to the β-form during storage was a key factor for the generation of granular crystals. Namely, a slow polymorphic transition rate to the β-form produced larger granular crystals during storage. Rapid
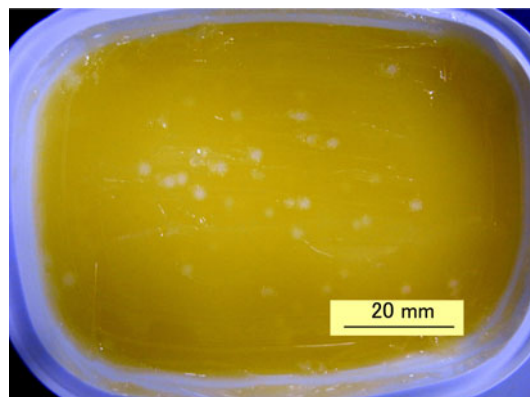

Fig. 9 Granular crystals observed in the margarine tub. Bulk fat blend sample with added oleic acid monoglyceride without shear, stored under thermal thawing between 5 and 20 °C every 12 h

formation to the β-form of palm oil-based blends either by adding saturated monoglycerides or by shearing prevented the generation of granular crystals, leading to the deterioration of quality during storage.

Conclusions

Almost all commercial margarines use monoglycerides as emulsifiers to stabilize the W/O emulsions. We investigated the effects of the fatty acid components of monoglycerides on the polymorph behavior during storage. The peak intensities of the β-polymorph of the samples with added saturated monoglycerides were stronger than those of the unsaturated monoglycerides, and the peak intensities of the β-form of the W/O fat blend emulsions were stronger those of the bulk fat blends. Applied shear promoted the polymorphic transitions to the β-form during storage of the W/O emulsion fat blends. In contrast, shear retarded those of the bulk fat blends. These results suggested that fatty acids combined with the monoglycerides on the surface of the water droplets influenced the polymorphic transformation of the fat crystal network in the continuous semi-solid fat phase because of the acyl–acyl interactions between the fatty acid residues of the monoglycerides and triacylglycerols in the fat crystal network. In general, the fat crystal bonds in semi-solid foods such as margarine are divided into primary bonds, which are strong and are generated by the rapid cooling process, and secondary bonds, which are much weaker and are generated by pin working. The polymorphic transitions to the β-form during storage of the W/O emulsion fat blends were promoted by applied shear, while those of the bulk fat blends were retarded. These results in this study provided important information for the selection of emulsifiers and the pin working conditions for controlling the growth of granular

crystals in industrial processes, especially for low *trans* fatty acid-type margarines and shortenings.

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