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STRUCTURE OF MARGARINES MADE WITH LOW ERUCIC ACID RAPESEED OIL

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Abstract

Margarine based on hydrogenated low erucic acid rapeseed oil shows a strong tendency to produce an unacceptable grainy structure. This is caused by rapid transition of the β'-form into the β-form of fat crystals. Sorbitan esters, monoacylglycerols, and a blend of acylglycerols were investigated as possible β'-stabilizers. Polymorphic transitions were evaluated by visual assessment, examination by polarized light microscopy, and x-ray diffraction analysis in margarines prepared on a laboratory scale and containing varying amounts of stabilizers. The evaluation was extended over a 7-week period of storage at 10°C. Sorbitan esters of fatty acids were found to be the most efficient stabilizers. If the stabilizer content was 0.4% (w/w) or more, the transition was retarded and the margarine retained its smooth texture throughout the storage period.

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Key Words: Emulsifiers, Fat crystals, Light microscopy, Margarine, Microstructure, Polymorphism, Rapeseed oil, Sorbitan esters, Stabilizers, x-Ray diffraction.

Introduction

The texture of margarine is important to the customer as well as to other end users in the food industry and food preparation establishments. deMan *et al.* (1990) defined texture as the external manifestation of structure and composition. Texture of margarine usually comprises properties such as hardness, spreadability, graininess, brittleness, oiliness, and stickiness (deMan, 1961, 1983). Changes in textural properties have been traced to polymorphic transformations of fat crystals. deMan *et al.* (1990) commented that relatively large β-crystals rather than smaller β'-crystals are found in grainy margarines; polymorphic transformations can lead to graininess (Juriaanse and Heertje, 1988).

Polymorphism of fats, especially pure triacylglycerols, has been studied extensively, *e.g.,* by Aronhime *et al.* (1990), Chawla *et al.* (1990), Garti and Sato (1986), Garti *et al.* (1986), Gibon *et al.* (1986), Hagemann and Rothfus (1988), Hernqvist (1988), Hernqvist and Larsson (1982), Larsson (1972), Lutton (1950, 1960, 1972), Reddy and Prabhakar (1096), and Sato and Kuroda (1987). Triacylglycerols can crystallize in four different modifications: sub-α, α, β', and β, characterized by a particular carbon chain packing and specific thermal stability. Transitions, which can occur between these forms, are in principle irreversible ($\alpha \rightarrow \beta' \rightarrow \beta$), except for the sub-$\alpha \leftrightarrow \alpha$ transition, which seems to be reversible (Gibon *et al.*, 1986). For products such as margarine, the $\beta' \rightarrow \beta$ transition is most important.

Natural fats and hydrogenated oils have been reported as having β'- or β-tendencies, according to the crystal structures which they tend to form. Naguib-Mostafa and deMan (1985), Rivarola *et al.* (1987), and Wiedermann (1978) have reported that factors, which are responsible for β'- or β-crystallization in fats, include their palmitic acid content, the distribution of the fatty acids on the glycerol moiety, and the trans-fatty acid and erucic acid contents. Natural fats such as lard and cocoa butter, and hydrogenated coconut, corn, olive, palm kernel, peanut, soybean, and sunflower oils are β-tending fats, whereas natural fats such as milkfat and tallow, and hydrogenated cottonseed, palm, and rapeseed oils crystallize in the β'-form.

Margarine formulated with β'-tending fats contains tiny needle-shaped crystals (2-5 μm in diameter). They are evenly dispersed in the fat phase and are able to retain the water phase. Fat crystallizes in a three-dimensional network which is capable of immobilizing a large amount of liquid oil (Juriaanse and Heertje, 1988, Rivarola *et al.*, 1987). On the other hand, margarine formulated with β-tending fats contains large crystals in the range of 5 to 25 μm (Askinazi *et al.*, 1987, deMan *et al.*, 1990, Juriaanse and Heertje, 1988, Naguib-Mostafa and deMan, 1985, Rivarola *et al.*, 1987). The crystals may impart a grainy or sandy texture and cause the product to be hard and brittle. Such crystals form a coarse three-dimensional structure and this can lead to the separation of the oil phase, a phenomenon known as 'oiling off' (Grinsted Products, 1986).

Hydrogenated rapeseed oil crystallizes in the relatively stable β'-form (Hernqvist and Anjou, 1983). Several years ago, the erucic acid content of most rapeseed grown throughout the world including Czecho-Slovakia, had been reduced from about 55% (w/w) to about 1% (w/w) through selective plant breeding. The oil produced from this cultivar is of good quality. It is labeled as 'low erucic acid rapeseed oil' (LEARO) in Czecho-Slovakia, LOBRA in Sweden, and Canola in Canada and the U.S.A. Hydrogenated LEARO has a strong tendency to transform from the β'-form to the β-form. Margarine containing hydrogenated LEARO may, on storage, change to a product which would be unacceptable because of its texture which may be perceived as sandy in the mouth (Hernqvist and Anjou, 1983). Such margarine may also cause problems in the bakery and confectionery industries.

For this reason, Hernqvist and Anjou (1983) examined the effect of diacylglycerols on the $\beta' \rightarrow \beta$-transition in hydrogenated LOBRA. Also Askinazi *et al.* (1987) reported that the $\beta' \rightarrow \beta$-transition in hydrogenated sunflower oil can be retarded by addition of monoacylglycerols. Grinsted Products (1986) recommended sorbitan esters as a β'-stabilizer of hydrogenated sunflower oil. Finally, Lee and deMan (1984) examined the effect of a number of surface-active lipids on the polymorphic behavior of hydrogenated canola oil.

The objective of the present work was to study the stabilization of the β'-form in margarine, formulated with hydrogenated LEARO, using monoacylglycerols, a blend of monoacylglycerols and diacylglycerols, and sorbitan esters as β'-stabilizers.

Materials and Methods

Sample preparation

Studies were carried out on the fat phase of margarines as well as margarines which were prepared on a laboratory scale.

The fat phase was a blend of non-selectively hydrogenated LEARO (94%, w/w) and a commercially refined liquid LEARO (6%, w/w). Hydrogenation was carried out on a commercial scale at Palma, Bratislava, Czecho-Slovakia (Krupp hydrogenator, 160°C, 0.2 MPa hydrogen pressure) using a nickel catalyst produced by STZ, Ústí nad Labem, Czecho-Slovakia, at a level of 0.3% of nickel in the oil (w/w). Physical and chemical characteristics and the fatty acid composition of the fats under study are presented in Table 1.

The margarine blends contained variable amounts of the fat phase (81-83%, w/w) depending on the addition of the stabilizer (0-2%, w/w), water phase (16%, w/w), emulsifier (Emulsifier C, 0.5%, w/w), and a small amount of salt, coloring, and flavoring. The margarines were produced in a 2-tube laboratory votator. Laboratory conditions simulated industrial production of margarine. Approx. 600 g of each margarine blend was melted at 50°C and emulsified for 20 min in the first tube (emulsifier). The blend was then quickly cooled to 8-10°C in the second tube (chiller), homogenized, and packaged. The margarine samples were stored at 10°C for 7 weeks from the day of production.

Czecho-Slovak legislation for margarine allows only Emulsifier C to be used as the emulsifying agent at a level of 0.5% (w/w).

The additives tested were:

(a) Emulsifier C (EC) - a blend consisting of approx. (w/w) 40% monoacylglycerols, 45% diacylglycerols, 10% triacylglycerols, and a small amount of free fatty acids and glycerol, commercially produced by STZ, Ústí nad Labem, Czecho-Slovakia.

(b) Dimodan CP (D) - distilled monoacylglycerols of fatty acids, commercially produced by Grinsted Products A/S, Brabrand, Denmark; U.S. Food, Drug, and Cosmetics Act, § 182.4505 (FAO/WHO, 1974).

(c) Famodan TS (F) - sorbitan esters of fatty acids commercially produced by Grinsted Products A/S, Brabrand, Denmark; U.S. Food, Drug, and Cosmetics Act, § 172.842 (as sorbitan monostearate) (FAO/WHO, 1974).

Table 1. Physical and chemical characteristics and fatty acid composition of the fats under study

Fat	Melting point [°C]	Iodine value [$cgI_2 \cdot g^{-1}$]	Trans acids [%, w/w]	Fatty acids 16:0	18:0	18:1	18:2	18:3	22:1
				[% (w/w)]					
Hydrog. LEARO	37.0	37.0	35.0	5.4	5.3	65.8	18.9	3.6	1.0
Liquid LEARO	--	112.0	--	5.4	1.5	57.0	24.3	10.8	1.0
Fat phase	36.7	87.6	34.6	5.4	5.0	46.0	19.0	3.7	1.0

Table 2. Physical and chemical characteristics and fatty acid composition of the additives tested

Fat	Melting point [°C]	Iodine value [$cgI_2 \cdot g^{-1}$]	ADI* [mg/kg/d]	14:0	16:0	18:0	18:1	18:2	18:3	20:1
				Fatty acid [% (w/w)]						
Emulsifier C	44.3	47.6	no limit	0.4	14.1	20.9	55.2	6.1	1.7	1.6
Famodan TS	57.0	5.5	0-25	1.4	45.0	51.3	0	1.6	0.7	0
Dimodan CP	50.1	76.0	no limit	0.7	25.3	3.5	31.2	38.3	1.0	0

* ADI = Acceptable Daily Intake for Man (mg/kg body mass/day) (FAO/WHO, 1974).

In our experiments, 0.5 to 2.0% (w/w) EC, 0.5% EC + 0.5 to 2.0% D, or 0.5% EC + 0.1 to 2.0% F were used. Physical and chemical characteristics and the fatty acid composition of the additives tested are presented in Table 2.

Tristearin (melting point 72.9°C) was obtained from Baker Chemicals N. V., Deventer, The Netherlands; it was recrystallized from acetone before the experiments and was used as a standard of the β-form crystals.

Physical and chemical characteristics

Melting points, trans-isomer contents, and iodine values were determined by AOCS methods (Walker, 1982) Cc 1-25, Cd 14-61, and Cd 1-25, respectively. Fatty acids were analyzed as their methyl esters by AOCS method Ce 1-62 using a Hewlett-Packard gas chromatograph.

Texture, microstructure, and polymorphic transitions

Texture of the fat phase and the margarines was examined visually. Microstructure of the samples was studied using a Zeiss Amplyval polarized light microscope. The fat was placed on a glass slide in a layer approximately 50 μm thick and then was pressed with a cover slip to form a thin film. Photomicrographs were taken on 100 ISO black-and-white 35-mm film.

Polymorphic forms of the fat crystals were determined by x-ray diffraction technique using a DRON UM1 diffractometer (Burevestnik, U.S.S.R.) with Cu Kα radiation, Ni-filtered, scanning velocity 1°/min. For the study of the $\beta' \rightarrow \beta$-transitions, the x-ray detector was scanned over diffraction angles in the range of 5 to 25° (2θ). The x-ray diffraction lines observed for triacylglycerols between 16° and 25° (2θ) are called 'short spacings' and are different for each polymorphic form. They correspond to the shortest distances between the fatty acid hydrocarbon chains (Gibon *et al.*, 1986, Rivarola *et al.*, 1987). Characteristic short spacings of the β'-form are 4.2×10^{-10} m (2θ around 21°) and 3.8×10^{-10} m (2θ around 23°); for the β-form, the short spacings are 4.6×10^{-10} m (2θ around 19°), 3.8×10^{-10} m (2θ around 23°), and 3.6×10^{-10} m (2θ around 24°) (Aronhime *et al.*, 1987, Gibon *et al.*, 1986, Naguib-Mostafa and deMan, 1985, Naguib-Mostafa *et al.*, 1985, Rivarola *et al.*, 1987, Sato and Kuroda, 1987). Triacylglycerols may occur as either double or triple hydrocarbon chain layers; this can be decided by the 'long spacing' lines occurring around 5° (2θ) (Gibon *et al.*, 1986, Rivarola *et al.*, 1987).

Results and Discussion

Texture

Visual examination of the samples immediately after their production revealed that the margarines had, in general, smooth textures. Margarine containing only EC and margarine containing a blend of EC + D rapidly acquired a grainy texture upon storage. In contrast, margarine which contained 0.4% (w/w) or more F had a smooth texture throughout the storage period.

Microstructure

The photomicrographs of hydrogenated LEARO, which crystallized spontaneously, show clusters of well-defined large spherulites (Fig. 1). They are characterized by long needles which radiate from a central point and attain sizes up to 25 μm (Hojerová, 1991). According to Naguib-Mostafa *et al.* (1985) and Rivarola *et al.* (1987), this morphology is characteristic of β-form triacylglycerols. Microscopic examination after storage showed noticeable differences in the crystalline structures both in the fat phase and in the margarines. Polarized light microscopy of samples which contained only EC showed marked variation in size and structure of the crystals during the storage test. Clumps of large crystals were characteristic of the fat phase

Fig. 1. *Hydrogenated low erucic acid rapeseed oil as viewed under a polarized light microscope.*
Fat crystals (β-polymorphic form) formed on spontaneous crystallization.

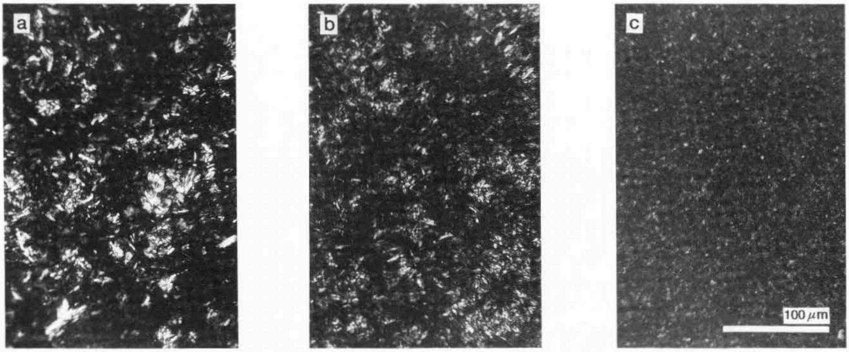

Fig. 2. Polarized light photomicrographs of fat crystals in margarine after 24 days of storage at 10°C.
***a:** grainy structure of margarine containing 0.5 % (w/w) of Emulsifier C; **b:** margarine containing 0.5% (w/w) of Emulsifier C + 0.5% (w/w) of Dimodan CP; **c:** smooth structure of margarine containing 0.5% (w/w) of Emulsifier C + 0.5% (w/w) Famodan TS. Fat crystals appear as light structures on a dark background.*

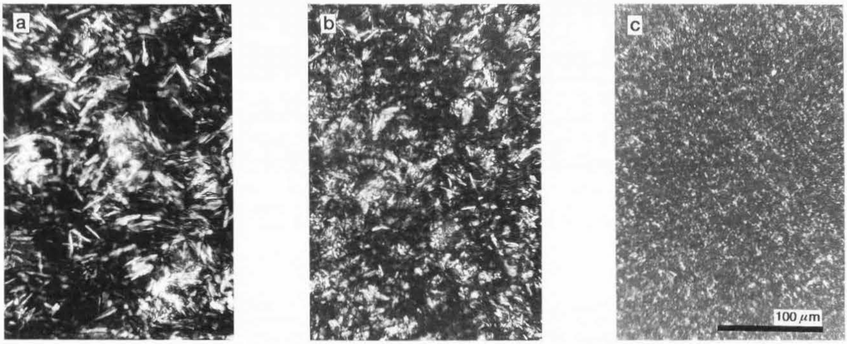

Fig. 3. Polarized light photomicrographs of fat crystals in margarine after 49 of storage at 10°C.
***a:** very grainy structure of margarine containing 0.5% (w/w) of Emulsifier C; **b:** margarine containing 0.5% (w/w) of Emulsifier C + 0.5% (w/w) of Dimodan CP; **c:** smooth structure of margarine containing 0.5% (w/w) of Emulsifier C + 0.5% (w/w) of Famodan TS.*

as well as the margarine prepared from it. The post-crystallization state of these margarines during storage is illustrated by Figs. 2a and 3a. This type of crystals and variations of their sizes are also present, though to a lesser degree, in the fat phase and in the margarine containing EC + D (Figs. 2b and 3b). This type of crystals was practically nonexistent in the margarine containing EC + F, where the proportion of F was 0.4% (w/w) or greater. The fat phase of this margarine solidified in the form of uniform small crystalline clusters. This morphology is characteristic of β'-form triacylglycerols (Naguib-Mostafa *et al.*, 1985, Rivarola *et al.*, 1987). The margarine with F (0.4% (w/w) or more) had a smooth texture and a fine crystalline structure throughout the entire storage period (Figs. 2c and 3c).

Polymorphic transition

A characteristic x-ray diffraction pattern of standard tristearin is shown in Fig. 4. Peaks at $4.6x10^{-10}$ m, $3.8x10^{-10}$ m, and $3.6x10^{-10}$ m can be seen in the tristearin pattern; they correspond to the β-form. Polymorphic forms of crystals present in the fat phase (hydrogenated LEARO) during storage at 10°C are characterized in Table 3 and polymorphic forms of crystals present in margarine (made with hydrogenated LEARO) during storage at 10°C are characterized in Table 4, where the weaker polymorphic form is shown in parentheses. Fig. 5 shows x-ray diffraction patterns of margarine after 13 days of storage at 10°C. The lower pattern in Fig. 5a (margarine containing 0.5% (w/w)

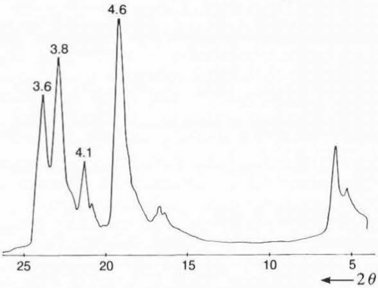

Fig. 4. Characteristic x-ray diffraction pattern of standard tristearin in β-form. Spacing $d = x \cdot 10^{-10}$ m; 2θ = Bragg diffraction angle.

Table 3. Polymorphic forms of crystals present in the fat phase (hydrogenated low erucic acid rapeseed oil) during storage at 10°C

Level of additives in the fat phase [%, w/w]	Polymorphic forms of crystals after storage at 10°C for 2 to 16 days			
	2 d	6 d	11 d	16 d
No additive	$(\beta')^* + \beta$	β	β	β
0.5 EC	$(\beta') + \beta$	β	β	β
1.0 EC	$\beta' + \beta$	β	β	β
1.5 EC	$\beta' + \beta$	$(\beta') + \beta$	β	β
2.0 EC	$\beta' + \beta$	$\beta' + \beta$	β	β
0.5 EC+ 0.5 D	$(\beta') + \beta$	β	β	β
0.5 EC+ 1.0 D	$(\beta') + \beta$	β	β	β
0.5 EC + 1.5 D	$(\beta') + \beta$	β	β	β
0.5 EC+ 2.0 D	$(\beta') + \beta$	β	β	β
0.5 EC+ 0.5 F	β'	β'	β'	β'
0.5 EC+ 1.0 F	β'	β'	β'	β'
0.5 EC+ 1.5 F	β'	β'	β'	β'
0.5 EC+ 2.0 F	β'	β'	β'	β'

EC = Emulsifier C; D = Dimodan CP; F = Famodan TS; * = Weaker polymorphic form is shown in parentheses.

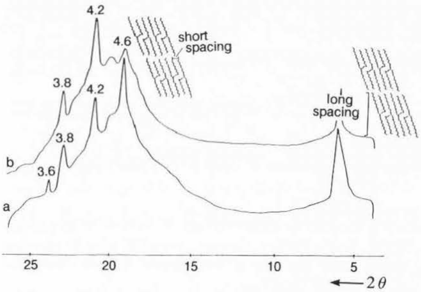

*Fig. 5. Characteristic x-ray diffraction patterns - spacing $d = x \cdot 10^{-10}$ m of margarine after 13 days of storage at 10°C. **a**: margarine containing 0.5% (w/w) of Emulsifier C (dominant β-form); **b**: margarine containing 0.5% (w/w) of Emulsifier C + 0.5% (w/w) of Famodan TS (dominant β'-form). 2θ = Bragg diffraction angle.*

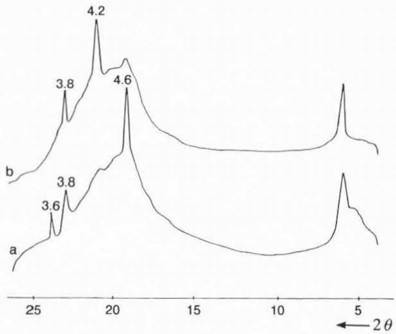

*Fig. 6. Characteristic x-ray diffraction patterns - spacing $d = x \cdot 10^{-10}$ m of margarine after 49 days of storage at 10°C. **a**: margarine containing 0.5% (w/w) of Emulsifier C (very dominant β-form); **b**: margarine containing 0.5% (w/w) Emulsifier C + 0.5% (w/w) Famodan TS (dominant β'-form). 2θ = Bragg diffraction angle.*

Table 4. Polymorphic forms of fat crystals in margarine, made with hydrogenated low erucic acid rapeseed oil and additives, during storage at 10°C for 4 to 49 days

Additives [%, w/w]	Polymorphic forms of crystals after storage 4 d	8 d	13 d	24 d	31 d	36 d	49 d
0.5 EC	$\beta'+\beta$	$(\beta')^*+\beta$	$(\beta')+\beta$	β	β	β	β
0.5 EC+ 0.1 F	β'	β'	$\beta'+(\beta)$	β	β	β	β
0.5 EC+ 0.2 F	β'	β'	β'	$\beta'+(\beta)$	$\beta'+\beta$	$(\beta')+\beta$	β
0.5 EC+ 0.3 F	β'	β'	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+\beta$	$\beta'+\beta$	$\beta'+\beta$
0.5 EC+ 0.4 F	β'	β'	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$
0.5 EC+ 0.5 F	β'	β'	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$	$\beta'+(\beta)$
0.5 EC+ 0.8 F	β'	β'	β'	β'	β'	β'	β'
0.5 EC+ 1.0 F	β'	β'	β'	β'	β'	β'	β'

EC = Emulsifier C; F = Famodan TS; * = Weaker polymorphic form is shown in parentheses.

EC) shows a strong peak at 4.6×10^{-10} m, which is characteristic of the β-form, and a weaker peak at 4.2×10^{-10} m, which is characteristic of the β'-form. Unlike the lower pattern, the upper pattern in Fig. 5 b (margarine containing 0.5% (w/w) EC + 0.5% (w/w) F) has a strong peak at 4.2×10^{-10} m and a weaker peak at 4.6×10^{-10} m. Fig. 6 shows x-ray diffraction patterns of margarine after 49 days of storage at 10°C. In the lower pattern (Fig. 6a: margarine containing 0.5% (w/w) EC), peaks at 4.6×10^{-10} m, 3.8×10^{-10} m, and 3.6×10^{-10} m can be seen; they correspond to the β-form. The upper pattern (Fig. 6b: margarine containing 0.5% (w/w) EC + 0.5% (w/w) F) showed constantly a strong peak at 4.2×10^{-10} m which is characteristic of the β'-form.

x-Ray diffraction patterns of all the samples as well as polarized light microscopy and visual judgment of the margarines showed that the Famodan TS sorbitan ester of fatty acids is an effective stabilizer of β'-crystals. Transition into the β-form was retarded and the margarine had a smooth texture throughout the storage period provided that the stabilizer concentration was 0.4% (w/w) or more.

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Discussion with Reviewers

D. M. Manning: All evaluations have been conducted on a combination of emulsifiers. Is there a reason for using combinations rather than individual emulsifier systems?

J. M. deMan: Do you expect 0.4% Famodan TS to have a similar stabilizing effect in the absence of Emulsifier C?

Authors: Famodan TS is not an emulsifying agent suitable for a margarine water-in-oil emulsion. It has a weak emulsifying effect only on oil-in-water emulsions (Grinsted Products, 1986).

Obviously, Famodan TS is an effective β'-stabilizer in hydrogenated canola oil also in the absence of an emulsifier (our previous unpublished results and Lee and deMan, 1984).

D. M. Manning: What is the scientific basis for Famodan TS being the preferred emulsifier/stabilizer?

Authors: The mechanism of action of surfactants on the $\beta' \rightarrow \beta$ transition is not well understood. According to Larsson (1972) and Lee and deMan (1984), the mechanism of polymorphic transition of β'-packing to β-packing would involve 180° rotation of the glyceride chains. Some surfactants, *e.g.,* sorbitan tristearate, are incorporated into the crystall lattice and block this rotation. To be effective, the surfactants should have the ability to co-crystallize with the fat.

J. M. deMan: It has been shown that the most effective way of stabilizing the β'-form in canola margarine is the use of palm oil or, preferrably, hydrogenated palm olein. Have you tried this?

Authors: We know of several studies on the addition of palm oil, its fraction, or cottonseed oil to canola margarine in order to stabilize it, but we did not study such oils.

Reviewer 3: Has any attempt been made to vary the rates of cooling to modify the initial crystal form and/or temper the products to find if specific storage temperatures provide any interactive effect?

Why was 10°C chosen for the product storage temperature since normal refrigerated storage for margarine products at retail or in home is 4°C?

Authors: The rate of cooling has a great influence on the crystallization of fats. In our previous study (Hojerová, 1991), we have shown crystalline morphologies obtained using slow and rapid crystallization of hydrogenated LEARO. The rate of cooling of margarine under industrial conditions is controlled by automated machinery. In our opinion, the storage temperature of margarine is a more difficult problem. The higher the temperature, the more rapid the transformation of the crystals from β' to β. The samples were therefore maintained at the highest temperature, *i.e.,* 10°C, which is permitted for storage of this type of margarine in Czecho-Slovakia.

D. M. Manning: In general, refrigerated products are regularly temperature-cycled by removing from refrigeration and then returning them into the refrigerator. How will this cycling influence the quality of stabilization?

Authors: We have found that the stabilizer plays an important role in the polymorphic transformation, but certainly other aspects, such as the processing conditions, must also be considered. The cycling of the storage temperature is assumed to lead to the appearance of the more stable polymorphic form (Lee and deMan (1984). The margarine should preferably be stored in cold stores, transported in refrigerated trucks, and sold from cold counters.

D. M. Manning: Is the grainy fat structure created by crystal transition perceivable to the average consumer? And is this a common complaint in the margarine industry?
Authors: The grainy structure is characteristic of margarines made exlusively with β-tending fats. The sandy mouthfeel is easily perceivable to the average consumer. The substantial changes in textural characteristics may also cause that the margarine is unusable, *e.g.,* in the bakery industry.

D. M. Manning: Figures 2 and 3 present micrographs of thin sample films. How was the pressing conducted in order to make these thin films? How would different pressures affect the crystals or crystal fragments observed in the micrographs? Approximately, what is the percent crystallinity on the micrographs and how does that compare to the original margarine sample?

Was a temperature-controlled microscope stage used to maintain the 10°C sample temperature during pressing and microscopic examination?
Authors: We tempered the glass slide and the cover slip beforehand at 10°C because our microscope lacked the thermoregulated stage. The cover slip, which was very light, also protected the microscope objective. Some measurements were conducted without the cover slip in place and there were no changes observed compared with measurements with the cover slip. This technique was also used by others, *e.g.,* Rivarola *et al.* (1987). The photomicrographs show the shapes and sizes of typical crystals in the margarine. We did not study the comprehensive crystallinity, *e.g.,* the number of crystals per mm^3 of the margarine.

Reviewer 3: Have the investigators studied the long-term stability of the products made? Seven weeks is a relatively short time from a marketing standpoint.
Authors: The microstructure of margarine was also studied following a 12-week storage but there were no differences observed compared with the measurements obtained after the 7-week storage. For this reason, the x-ray diffraction measurements on the margarine samples were carried out after the 7-week storage only.